Optical tristability and ultrafast Fano switching in nonlinear magnetoplasmonic nanoparticles

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We consider light scattering by a coated magnetoplasmonic nanoparticle with a Kerr-type nonlinear plasmonic shell and a magneto-optic core. Such a structure features two plasmon dipole modes, associated with electronic oscillations on the inner and outer surfaces of the shell. Driven in a nonlinear regime, each mode exhibits a bistable response. Bistability of an inner plasmon leads to switching between this state and a Fano resonance (Fano switching). Once the external light intensity exceeds a critical value, the bistability zones of both eigenmodes overlap, yielding optical tristability characterized by three stable steady states for a given wavelength and light intensity. We develop a dynamic theory of transitions between nonlinear steady states and estimate the characteristic switching time to be as short as 0.5 ps. We also show that the magneto-optical effect allows red and blue spectral shifts of the Fano profile for right and left circular polarizations of the external light circular polarization while cancelling it for the left circular polarization. The results point to a class of ultrafast Fano switchers tunable by a magnetic field for applications in nanophotonics.

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I. INTRODUCTION

Magnetoplasmonic architectures, consisting of plasmonic and magneto-optical (MO) elements, have recently become an active topic of research due to their multifunctionality stemming from magnetic tunability and nonreciprocity [1–7]. For instance, in magnetoplasmonic systems containing a metallic film perforated with subwavelength hole arrays and a dielectric film magnetized perpendicular to its plane, extraordinary transverse MO Faraday and Kerr effects were predicted [1]. Along this line, the giant enhancement of the transverse MO effect was experimentally found in magnetoplasmonic crystals composed of periodic gold nanowires structured on a thin layer of a MO dielectric [8,9]. Furthermore, Belotelov et al. demonstrated MO-induced modulation of the transparency for magnetoplasmonic crystals [10]. Based on the Lorentz nonreciprocal model, Floess et al. derived the analytical expressions for a resonantly enhanced MO response in hybrid magnetoplasmonics and unraveled the underlying interplay between waveguide-plasmon-polariton quasiparticles and the Faraday rotation [5]. Beyond that, a great deal of attention has been paid to plasmonic nanorods in a MO medium [6], magnetoplasmonic dimers [11,12], and magnetic nanoparticle arrays [13]. Additionally, a number of interesting effects has been predicted for core-shell magnetoplasmonic nanoparticles (MPNPs) such as MO spasers [14], tunable plasmonic cloaks [15], enhanced Faraday rotation [16,17], circular dichroism [18], as well as the plasmon-driven Hall photon currents [19].

As is known, a coated nanoparticle encompassing a dielectric core and a concentric spherical plasmonic shell possesses two dipole plasmonic eigenmodes related to electronic oscillations localized on the outer and inner shell surfaces [20]. Once they oscillate out of phase, the full particle dipole moment almost vanishes, resulting in a Fano spectral shape of the scattering cross section (because of that this mode is also referred to as dark) [21]. In general, the Fano resonance (FR) features a steep, asymmetric line shape and an inherently excellent sensitivity to the changes in geometry compared to conventional resonance modes [22,23]. Due to a large enhancement of the local fields, FR in a core-shell particle can boost the optical nonlinearity residing in a metallic shell and/or a dielectric core, as it has been shown for a variety of structures demonstrating a Kerr-type nonlinear response [24-30]. The local field growth was also exploited to reach the strong control over second-harmonic generation via the MO effect [31–33].

In this paper, we investigate the optical bistable/tristable behavior of coated MPNPs consisting of a MO core and a Kerr-type nonlinear plasmonic shell. In Sec. II we present the general theoretical model describing both the nonlinear steady states and dynamical switching between them. Being in the framework of a quasistatic approximation, we account for the intrinsic nonlinear response of the metal via the self-consistent mean-field approximation [34–36] and find the steady-state solution. Then we employ the dispersion relation method [37–42] to develop the dynamical theory of switching. In Sec. III we show that the bistability of plasmonic modes leads

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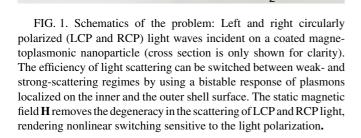
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to tristability and switching between the bright high-energy plasmon mode and the dark Fano mode. We demonstrate the temporal dynamics of such transitions and estimate the characteristic switching time. Finally, we predict MO-induced tunability of the bistable spectral domains for the particle scattering cross section. Specifically, red and blue spectral shifts of the Fano profile for right and left circular polarizations of the external light upon applying the external magnetic field are presented. Section IV summarizes the main results of the article.

II. THEORETICAL MODELS

Figure 1 illustrates the considered core-shell nanoparticle, a MO (Bi:YIG) core with the permittivity tensor $\vec{\varepsilon}_{c}$ and the radius r coated by a silver shell (the radius R > r) with the nonlinear permittivity $\tilde{\varepsilon}_s = \varepsilon_s + \chi^{(3)} |\mathbf{E}_s|^2$, where $\varepsilon_s = \varepsilon_{\infty} - \varepsilon_{\infty}$ $\omega_p^2/(\omega^2 + i\nu\omega)$ ($\varepsilon_{\infty} = 4.96$, $\hbar\omega_p = 9.54$ eV) is the linear permittivity, $\chi^{(3)} = 3 \times 10^{-9}$ esu is the typical cubic susceptibility of silver nanoparticles [43,44], and $|\mathbf{E}_s|^2$ the local intensity of the electric field inside the shell. The particle size is supposed to be much smaller than the light wavelength. Hereinafter we accept the harmonic time dependence $\exp(-i\omega t)$. We consider reduced losses $\hbar v = 0.0055$ eV (which can be achieved via decreasing the ambient temperature [45]) to get the pronounced Fano profile of the scattering cross section. Similar particles have been extensively studied both experimentally [16,46] and theoretically [17–19,47]. We assume that the MO core is magnetized by the external uniform magnetic field H along the z axis. Thus, the gyrotropy of the core is described by a relative permittivity tensor,

$$\vec{\varepsilon}_c = \begin{pmatrix} \varepsilon & ig & 0\\ -ig & \varepsilon & 0\\ 0 & 0 & \varepsilon \end{pmatrix}.$$



RCF

Here, the gyration $g = \chi_m H$ (χ_m is the magneto-optical susceptibility) is responsible for the "strength" of MO activity. We accept that the MO effect is induced by an external magnetic field only. For a typical MO-active material Bi:yttrium iron garnet (YIG), $\varepsilon = 5.5 + 0.0025i$ [1], and the value of g is tunable in the range of 0–0.3 by adjusting the magnetic field [48,49]. Since its 6-nm-thick films demonstrate $\chi_m \approx 3.8 \times 10^{-5}$ esu [50], the maximal value of gyration which can be reached in practice corresponds to a magnetic field of ~7.8 kG (or 0.78 T).

Note that here, for the sake of clarity, we ignore the metallic nonlocality which potentially may lead to a spectral shifting of the Fano resonance peak and enhancement of the nonlinear response [51].

A. Nonlinear steady-state solution

To begin with, we consider the linear problem (i.e., $\chi^{(3)} = 0$) within the quasistatic dipole approximation, supposing that the size of the nanoparticle is much smaller than the incident light wavelength. We look for the local electric field inside the core, the shell, and outside the nanoparticle in the form [14,52]

$$\mathbf{E}_{\text{core}} = \hat{A}\mathbf{E}_i,\tag{1a}$$

$$\mathbf{E}_{\text{shell}} = \hat{B}\mathbf{E}_i - \frac{\hat{C}\mathbf{E}_i}{\rho^3} + \frac{3(\hat{C}\mathbf{E}_i \cdot \mathbf{n})\mathbf{n}}{\rho^3}, \quad (1b)$$

$$\mathbf{E}_{\text{out}} = -\frac{\hat{D}\mathbf{E}_i}{\rho^3} + \frac{3(\hat{D}\mathbf{E}_i \cdot \mathbf{n})\mathbf{n}}{\rho^3} + \mathbf{E}_i, \qquad (1c)$$

where \mathbf{E}_i is the incident field, \hat{A} , \hat{B} , \hat{C} and \hat{D} are unknown antisymmetric tensors, and ρ is the distance from the particle geometric center. Having applied the boundary conditions for Eqs. (1a)–(1c) on the inner and outer surfaces, we arrive at the following system of equations,

$$(\varepsilon \hat{I} + \hat{G})\hat{A} = \varepsilon_s \hat{B} + 2\varepsilon_s \frac{\hat{C}}{r^3},$$
$$\varepsilon_s \hat{B} + 2\varepsilon_s \frac{\hat{C}}{R^3} = 2\frac{\hat{D}}{R^3} + \hat{I},$$
$$\hat{A} = \hat{B} - \frac{\hat{C}}{r^3},$$
$$\hat{B} - \frac{\hat{C}}{R^3} = -\frac{\hat{D}}{R^3} + \hat{I},$$

where \hat{G} is the nondiagonal part of the permittivity tensor $\vec{\varepsilon}_{c}$. These relations allow us to write the tensors \hat{A} , \hat{B} , \hat{C} , and \hat{D} in the same form,

$$\hat{M} = \begin{pmatrix} M_{11} & i M_{12} & 0\\ -i M_{12} & M_{11} & 0\\ 0 & 0 & M_{33} \end{pmatrix} \quad (M = A, B, C, D), \quad (2)$$

Metallic shell

NO core

where

$$\begin{split} A_{11} &= \frac{9R^{3}\varepsilon_{s}[2r^{3}(\varepsilon_{s}-1)(\varepsilon-\varepsilon_{s})+R^{3}(\varepsilon_{s}+2)(\varepsilon+2\varepsilon_{s})]}{P_{+}P_{-}}, \\ A_{12} &= \frac{-9gR^{3}\varepsilon_{s}[2r^{3}(\varepsilon_{s}-1)+R^{3}(\varepsilon_{s}+2)]}{P_{+}P_{-}}, \\ B_{11} &= \frac{3R^{3}[g^{2}[2r^{3}(1-\varepsilon_{s})-R^{3}(\varepsilon_{s}+2)]+(\varepsilon+2\varepsilon_{s})[2r^{3}(\varepsilon-\varepsilon_{s})(-1+\varepsilon_{s})+R^{3}(\varepsilon_{s}+2)(\varepsilon+2\varepsilon_{s})]]}{P_{+}P_{-}}, \\ B_{12} &= \frac{18gR^{3}r^{3}\varepsilon_{s}(1-\varepsilon_{s})}{P_{+}P_{-}}, \\ C_{11} &= \frac{3R^{3}r^{3}[g^{2}[2r^{3}(1-\varepsilon_{s})-R^{3}(\varepsilon_{s}+2)]+(\varepsilon-\varepsilon_{s})[2r^{3}(\varepsilon-\varepsilon_{s})(-1+\varepsilon_{s})+R^{3}(\varepsilon_{s}+2)(\varepsilon+2\varepsilon_{s})]]}{P_{+}P_{-}}, \\ C_{12} &= \frac{9gR^{6}r^{3}\varepsilon_{s}(2+\varepsilon_{s})}{P_{+}P_{-}}, \\ D_{11} &= \frac{R^{3}}{P_{+}P_{-}}[[R^{3}(\varepsilon+2\varepsilon_{s})(\varepsilon_{s}-1)+r^{3}(\varepsilon-\varepsilon_{s})(1+2\varepsilon_{s})][R^{3}(\varepsilon+2\varepsilon_{s})(\varepsilon_{s}+2)+2r^{3}(\varepsilon-\varepsilon_{s})(\varepsilon_{s}-1)]}{-g^{2}[R^{3}(\varepsilon_{s}-1)+r^{3}(1+2\varepsilon_{s})][R^{3}(\varepsilon_{s}+2)+2r^{3}(\varepsilon_{s}-1)]], \\ D_{12} &= \frac{27gR^{6}r^{3}\varepsilon_{s}^{2}}{P_{+}P_{-}}, \end{split}$$

with $P_{\pm} = -2r^3(\varepsilon - \varepsilon_s)(1 - \varepsilon_s) + R^3(2 + \varepsilon_s)(\varepsilon + 2\varepsilon_s) \pm g[2r^3(1 - \varepsilon_s) - R^3(2 + \varepsilon_s)]$. When $E_{iz} = 0$, there exist two eigenvalues for the tensor \hat{D} , i.e.,

$$\alpha_{\pm} = D_{11} \pm i D_{12} = R^3 \frac{r^3 (2\varepsilon_s + 1)(\varepsilon \mp g - \varepsilon_s) + R^3 (-1 + \varepsilon_s)(\varepsilon \mp g + 2\varepsilon_s)}{2r^3 (-1 + \varepsilon_s)(\varepsilon \mp g - \varepsilon_s) + R^3 (2 + \varepsilon_s)(\varepsilon \mp g + 2\varepsilon_s)},\tag{3}$$

where -g corresponds to $\mathbf{E}_{+} = (1 \quad i \quad 0)^{T}$ polarization, while +g to $\mathbf{E}_{-} = (1 \quad -i \quad 0)^{T}$, and α_{\pm} is the particle polarizability corresponding to left and right circularly polarized (LCP and RCP) light, respectively.

Next, we account for the Kerr-type nonlinearity of the metallic shell. In general, there is no analytical solution for such a problem as the local field in the shell is inhomogeneous, and one should solve numerically the nonlinear Laplace equation. However, when the nonlinear contribution is weak (i.e., $\varepsilon_s \gg \chi^{(3)} |\mathbf{E}_s|^2$), it can be considered in the first order of perturbation. This means that one can neglect nonlinearity-driven variance in the field structure and account for the self-action of the

electromagnetic field via the averaged field [34]. Hence, we present the shell permittivity as

$$\tilde{\varepsilon}_s = \varepsilon_s + \chi^{(3)} |\mathbf{E}_s|^2 \approx \varepsilon_s + \chi^{(3)} \langle |\mathbf{E}_s|^2 \rangle.$$
(4)

Here, the field averaged in the shell volume V_s can be expressed as

$$\langle |\mathbf{E}_s|^2 \rangle = \frac{1}{V_s} \int_r^R \int_0^\pi \int_0^{2\pi} \mathbf{E}_s \mathbf{E}_s^* \rho^2 \sin\theta d\rho d\theta d\phi, \quad (5)$$

and the local electric field can be written in accord with Eqs. (1b) and (2) as

$$\begin{split} \mathbf{E}_{s} &= \begin{pmatrix} B_{11}E_{ix} + B_{12}E_{iy} \\ -B_{12}E_{ix} + B_{11}E_{iy} \\ B_{33}E_{iz} \end{pmatrix} - \frac{1}{\rho^{3}} \begin{pmatrix} C_{11}E_{ix} + C_{12}E_{iy} \\ -C_{12}E_{ix} + C_{11}E_{iy} \\ C_{33}E_{iz} \end{pmatrix} \\ &+ \frac{3}{\rho^{3}} \begin{pmatrix} (C_{11}E_{ix} + C_{12}E_{iy})\sin\theta^{2}\cos\phi^{2} + (-C_{12}E_{ix} + C_{11}E_{iy})\sin\theta^{2}\sin\phi\cos\phi + C_{33}E_{iz}\sin\theta\cos\theta\cos\phi \\ (C_{11}E_{ix} + C_{12}E_{iy})\sin\theta^{2}\sin\phi\cos\phi + (-C_{12}E_{ix} + C_{11}E_{iy})\sin\theta^{2}\sin\phi^{2} + C_{33}E_{iz}\sin\theta\cos\theta\sin\phi \\ (C_{11}E_{ix} + C_{12}E_{iy})\sin\theta\cos\phi\cos\phi + (-C_{12}E_{ix} + C_{11}E_{iy})\sin\theta\cos\phi\sin\phi + C_{33}E_{iz}\cos\theta^{2} \end{pmatrix}, \end{split}$$

where θ and ϕ are spherical polar and azimuthal angles.

When the incident light is LCP or RCP, i.e., $\mathbf{E}_i = E_0 e^{ikz} (1 \pm i \ 0)^T$, Eq. (5) can be simplified by using the spectral representation method [25,53] and written as follows,

$$\langle |\mathbf{E}_{s}|^{2} \rangle = \frac{2E_{0}^{2}}{(2+\varepsilon\pm g)^{2}} \bigg(\frac{|X|^{2}}{|S-S_{1}|^{2}} + 2\eta^{3} \frac{|Y|^{2}}{|S-S_{2}|^{2}} \bigg), \quad (6)$$

where $\eta = r/R$ is the aspect ratio between the core and the shell and

$$S = \frac{1}{1 - \varepsilon_s - \chi^{(3)} \langle |\mathbf{E}_s|^2 \rangle},$$

$$X = \frac{S[S(\varepsilon \pm g + 2) - 2]}{S - S_2},$$

$$Y = \frac{S[S(\varepsilon \pm g - 1) + 1]}{S - S_1}.$$

The poles S_1 and S_2 are given by

$$S_{1} = \frac{b - \sqrt{b^{2} - 24(2 + \varepsilon \pm g)(1 - \eta^{3})}}{6(2 + \varepsilon \pm g)},$$

$$S_{2} = \frac{b + \sqrt{b^{2} - 24(2 + \varepsilon \pm g)(1 - \eta^{3})}}{6(2 + \varepsilon \pm g)},$$

with $b = 8 + \varepsilon \pm g + 2\eta^3 (\varepsilon \pm g - 1)$.

Now, to obtain the nonlinear particle polarizability, one should exchange ε_s with $\varepsilon_s + \chi^{(3)} \langle |\mathbf{E}_s|^2 \rangle$ in Eq. (3) so that Eqs. (3) and (6) form the self-consistent nonlinear steady-state solution.

B. Dynamical model

To characterize the temporal behavior of our system, we derive the dynamical model for the MPNP response to the

external electric field. To this end, we employ the dispersion relation method [37] adapted for plasmonic nanoparticles [38–41] and graphene flakes [42], and write the Fourier transforms of the MPNP electric-dipole moment as

$$\alpha_{\pm}^{-1}(\omega)\mathbf{p}_{\pm} = \mathbf{E}_i,\tag{7}$$

where the subscript "±" corresponds to LCP and RCP. Assuming that $\chi^{(3)}\langle |\mathbf{E}_s|^2 \rangle \ll 1$ and $\nu/\omega_0 \ll 1$, we decompose $\alpha_{\pm}^{-1}(\omega)$ in the vicinity of the mode eigenfrequencies $\omega_{1,2}$, and keep the first-order terms involving time derivatives to account for the small broadening of the MPNP polarization spectrum,

$$\alpha_{\pm}^{-1} = \alpha_{\pm}^{-1}(\omega_{1,2}) + \frac{\partial \alpha_{\pm}^{-1}}{\partial \omega} \bigg|_{\omega = \omega_{1,2}} \left(\Delta \omega_{1,2} + i \frac{d}{dt} \right), \quad (8)$$

where $\Delta \omega_{1,2} = \omega - \omega_{1,2}$ is the frequency shift from the resonance value. The nonlinear term in Eq. (8) should be expressed via the particle dipole moment to reach the self-consistent model. Therefore, we use Eq. (6) at $\chi^{(3)} = 0$ and $E_0 = \alpha_{\pm}^{-1}(\omega_{1,2})p_{\pm}$.

Next, we substitute Eq. (8) into Eq. (7) and obtain the governing dynamical equations for the slowly varying amplitudes of the particle polarization $P_{1,2}^{\pm}$ corresponding to the plasmon modes localized in the outer and inner surfaces of the silver shell,

$$i\frac{dP_{1}^{\pm}}{d\tau} + (i\gamma_{1} + \Omega_{1} + |P_{1}^{\pm}|^{2})P_{1}^{\pm} = E,$$

$$i\frac{\omega_{1}}{\omega_{2}}\frac{dP_{2}^{\pm}}{d\tau} + (i\gamma_{2} + \Omega_{2} + \varsigma^{2}|P_{2}^{\pm}|^{2})\varsigma P_{2}^{\pm} = \kappa E,$$
(9)

where $\tau = \omega_1 t$, $\Omega_{1,2} = \Delta \omega_{1,2}/\omega_{1,2}$, and $\gamma_{1,2} = \text{Im}\{\alpha_{\pm}^{-1}(\omega_{1,2})\}/(\partial_{\omega}\alpha_{\pm}^{-1}|_{\omega=\omega_{1,2}}\omega_{1,2})$. Here, we use the following normalization,

$$\begin{split} E &= E_{i}(\psi_{1}\xi_{1})^{1/2} \left(\partial_{\omega} \alpha_{\pm}^{-1} \big|_{\omega=\omega_{1}} \omega_{1}\right)^{-3/2}, \\ P_{1,2}^{\pm} &= p_{\pm 1,\pm 2} \Big[\psi_{1}\xi_{1} / \left(\partial_{\omega} \alpha_{\pm}^{-1} \big|_{\omega=\omega_{1}} \omega_{1}\right)\Big]^{1/2}, \\ \psi_{1,2} &= \frac{\left[2r^{3}(\varepsilon \mp g + 1 - 2\varepsilon_{s}) + R^{3}(4 + \varepsilon \mp g + 4\varepsilon_{s})\right]\chi^{(3)}}{\left[r^{3}(2\varepsilon_{s} + 1)(\varepsilon \mp g - \varepsilon_{s}) + R^{3}(\varepsilon \mp g + 2\varepsilon_{s})(\varepsilon_{s} - 1)\right]R^{3}}\Big|_{\substack{\omega=\omega_{1,2}\\ Im\{\varepsilon_{s}\}=0\\ Im\{\varepsilon\}=0}}, \\ \xi_{1,2} &= \frac{18R^{3}[2r^{3}(\varepsilon \mp g - \varepsilon_{s})^{2} + R^{3}(\varepsilon \mp g + 2\varepsilon_{s})^{2}]}{\left([r^{3}(2\varepsilon_{s} + 1)(\varepsilon \mp g - \varepsilon_{s}) + R^{3}(-1 + \varepsilon_{s})(\varepsilon \mp g + 2\varepsilon_{s})]R^{3}\right)^{2}}\Big|_{\substack{\omega=\omega_{1,2}\\ Im\{\varepsilon_{s}\}=0\\ Im\{\varepsilon_{s}\}=0}} \end{split}$$

The terms

$$\kappa = \frac{(\psi_{2}\xi_{2})^{1/2} (\partial_{\omega}\alpha_{\pm}^{-1}|_{\omega=\omega_{2}}\omega_{2})^{-3/2}}{(\psi_{1}\xi_{1})^{1/2} (\partial_{\omega}\alpha_{\pm}^{-1}|_{\omega=\omega_{1}}\omega_{1})^{-3/2}},$$

$$\varsigma = \left(\frac{\psi_{1}\xi_{1}}{\partial_{\omega}\alpha_{\pm}^{-1}|_{\omega=\omega_{1}}\omega_{1}}\right)^{-1/2} \left(\frac{\psi_{2}\xi_{2}}{\partial_{\omega}\alpha_{\pm}^{-1}|_{\omega=\omega_{2}}\omega_{2}}\right)^{1/2}$$

characterize the system dispersion.

Finally, to get the full system response, one should summarize solutions of Eq. (9), i.e., $P^{\pm} = P_1^{\pm} + P_2^{\pm}$. For example, the steady-state solution is given by

$$(i\gamma_{1} + \Omega_{1} + |P_{1}^{\pm}|^{2})P_{1}^{\pm} = E,$$

$$(i\gamma_{2} + \Omega_{2} + \varsigma^{2}|P_{2}^{\pm}|^{2})\varsigma P_{2}^{\pm} = \kappa E.$$
 (10)

Being physically identical to the nonlinear solution presented by Eqs. (3) and (6), Eq. (10) describes the Fano resonance, bistability, multistability, as well as MO tuning of the nonlinear properties.

III. RESULTS AND DISCUSSION

A. Analysis of the nonlinear steady-state solution

To provide a quantitative characterization for the light-MPNP interaction, we introduce the light scattering efficiency which is defined as $Q_{\rm sc} = 8/3(2\pi R/\lambda)^4 |\Delta_1|^2$, with $\Delta_1 = \alpha_{\pm}/R^3$ [54], where α_{\pm} can be given by Eqs. (3) and (6) or by $\alpha_{\pm} = (P^{\pm}/E)[\partial_{\omega}\alpha_{\pm}^{-1}|_{\omega=\omega_1}\omega_1]^{-1}$.

First, we consider a linear nonmagnetic regime. Figure 2(a) plots the electric field structure for the different modes of the MPNP. There exist two bright dipole resonant modes, associated with electronic oscillations on the surfaces of the shell (outer and inner plasmons), and the dark Fano mode has arisen as a result of out-of-phase oscillations of these modes. The relative spectral position of the mode resonant wavelengths depends on the ratio between permittivities of the core and the host medium [51]. Since in our case $\varepsilon > 4\varepsilon_h$ ($\varepsilon_h = 1$), the eigenwavelength of the spherelike plasmon localized on the outer surface is smaller than the one of the cavitylike plasmon localized on the inner surface ($\lambda_s < \lambda_c$) [Figs. 2(b) and 2(c)]. The aspect ratio defines the strength of mode coupling evidenced by the eigenwavelengths splitting. For $\eta \leq$ 0.1 (the thick shell), the plasmons are almost decoupled and the lines of the Fano mode and the inner plasmon approach each other, yielding a narrow-band Fano-shape curve [51]. As the shell gets thinner, the mode interaction grows, leading to strong splitting of the mode wavelengths. It would be instructive to note that in the case $\varepsilon < 4\varepsilon_h$, the Fano-shape profile appears,

in sharp contrast to our system, in the vicinity of the outer plasmon [21].

Next, we investigate the impact of Kerr-type nonlinearity on the scattering efficiency still in a nonmagnetic regime. As the light intensity grows, nonlinearity first gives rise to bistability for the outer plasmon dipole mode along with an almost zero effect on the Fano resonance [see Figs. 2(d) and 2(e)]. Importantly, such behavior can be reached for comparatively low optical fields ~20 statvolt/cm (6×10^5 V/m) due to the synergy of the resonance local field enhancement and the high cubic metallic susceptibility.

It is methodologically useful to compare solutions given by the direct averaging and the dispersion relation method. They are shown in Figs. 2(c)-2(e). Remarkably, since both these techniques work in the first-order perturbation theory, they yielded almost identical results.

Once E_0 exceeds the critical value ~50 statvolt/cm (1.5 × 10⁶ V/m), both dipole modes demonstrate optical bistable regions (OBRs) [Fig. 3(a)]. Additionally, one can undergo a transition between the bright high-energy dipole mode and the dark Fano mode once the bistability domain is wide enough. For stronger fields, OBRs get overlapped, yielding an optical tristable region (OTR) or optical tristability characterized by three stable steady states [see Fig. 3(b)]. These states correspond to resonant dipole plasmons and an off-resonance response. The appearance of two separate OBRs and OTR can also be observed in Figs. 3(c) and 3(d), which show the

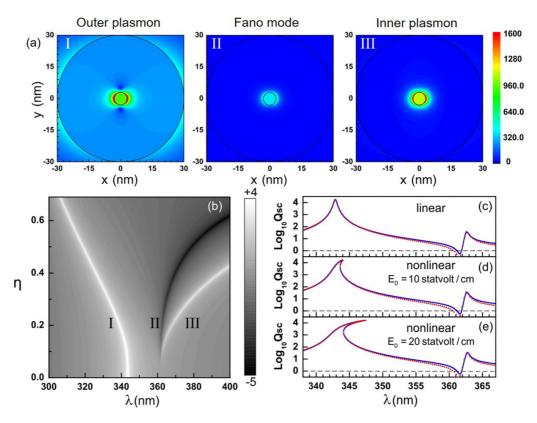


FIG. 2. (a) The electric field structure for the eigenmodes and the Fano mode of the core-shell particle when $\eta = 0.1$. Mode labels correspond to (b). (b) Linear light scattering efficiency as a function of wavelength and aspect ratio in the logarithmic scale $[\log_{10}(Q_{sc})]$. (c)–(e) Scattering efficiency spectra at different external fields for $\eta = 0.1$ and R = 30 nm. Continuous blue and dotted red curves correspond to solutions given by the direct averaging [Eqs. (3) and (6)] and the dispersion relation method [Eq. (10)].

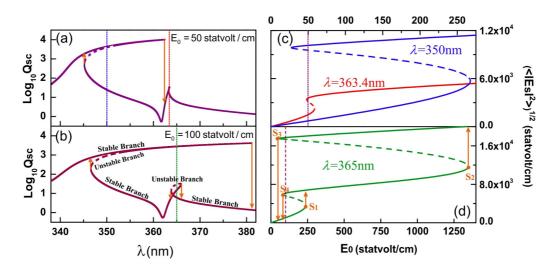


FIG. 3. (a), (b) Scattering efficiency spectra in the nonlinear regime for different values of the driving electric field. (c), (d) The average optical field in the shell as a function of the incident field for different wavelengths indicated in (a) and (b) by dashed lines. Vertical dashed lines indicate the fields for which (a) and (b) were plotted. All other parameters are the same as in Fig. 2.

average field in the shell versus the driving field E_0 for different wavelengths.

Importantly, the resonant enhancement of the local field inside the shell may reach a factor of several hundreds relative to the incident field, which is shown in Fig. 4(a). This allows us to reach OBRs and OTR at a moderate optical field facilitating the usage of the proposed core-shell nanoparticles as nonlinear nanoswitchers. We also point out that OTR can be obtained only for a wavelength larger than the eigenwavelength of the inner plasmon mode. This stems from the fact that Kerr-type metallic nonlinearity has a focusing type resulting in a redshift of the eigenwavelengths as the optical intensity grows, as shown in Fig. 4(a).

Let us now analyze the impact of shell thickness on bistability and multistability. To this end, we plot the bifurcation diagram showing bistable and tristable regions for a fixed wavelength near the Fano resonance, as a function of the volume fraction η and the incident field E_0 in Fig. 4(b). When the shell is significantly thicker than the core ($0 < \eta \le 0.137$), the plasmons are weakly interacting and their eigenfrequencies are not shifting as η grows [Fig. 2(b)]. In this situation, one can observe a clear Fano resonance and overlapping OBRs of both dipole modes, leading to tristability. For example, at $\eta = 0.1$ [dashed line in Fig. 4(b)], the threshold fields are consistent with the switching points S_1 - S_4 marked in Fig. 3(d). However, decreasing the shell thickness gives rise to strong plasmon hybridization, which shifts eigenfrequencies from each other [Fig. 2(b)]. As a result, one cannot reach OTR at a realistic optical power.

Next, we analyze tuning the scattering efficiency by the gyrotropy of the core. The MO effect exerts pronounced shifts in the eigenwavelength of the inner plasmon, while having almost no impact on its counterpart [Fig. 5(a)]. Hence, the associated Fano line experiences blueshifts and redshifts for LCP and RCP waves, respectively.

Figures 5(b) and 5(c) illustrate the impact of gyrotropy on optical bistability and multistability. In absence of \mathbf{H} (g = 0),

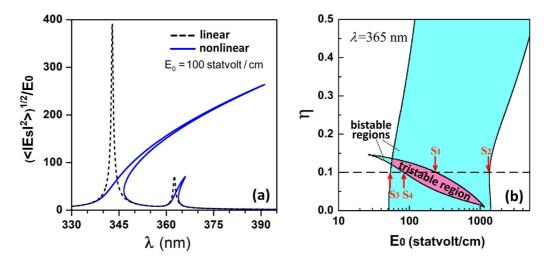


FIG. 4. (a) The enhancement factor $\langle |E_s|^2 \rangle^{1/2} / E_0$ vs the wavelength in the linear and nonlinear regimes. (b) Bifurcation diagram showing bistability and tristability zones as functions of volume fraction η and the incident field E_0 for $\lambda = 365$ nm. The points $S_1 - S_4$ correspond to the bistability thresholds marked in Fig. 3(d).

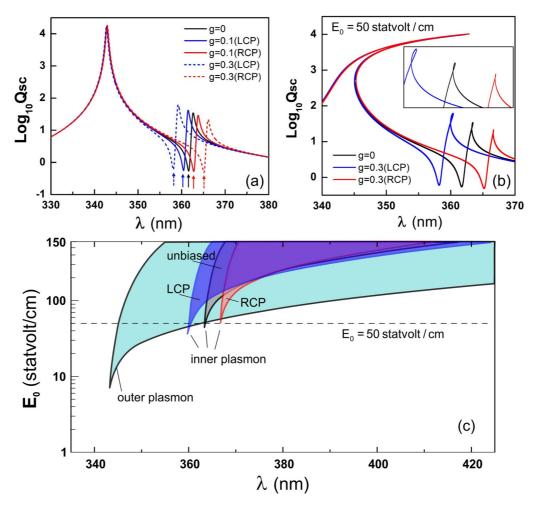


FIG. 5. (a) Linear and (b) nonlinear scattering efficiency spectra for right and left circularly polarized light in the presence of the MO effect. The inset shows the resonance lines for the inner plasmon in detail. (c) Bistability/tristability zones in (E_0,λ) axes for nonmagnetic and magnetic (g = 0.3) regimes. Magnetization just slightly shifts the bistability zone for the outer plasmon (unnoticeable in a logarithmic scale). For all figures, $\eta = 0.1$.

LCP and RCP light yield an equal scattering efficiency [the same as in Fig. 3(a)]. Although the MO effect almost does not influence the bistability zone of the outer plasmon, the Fano line and the bistability domain of the inner plasmon get considerably blue- and redshifted for the LCP and RCP wave. This, in particular, yields an interesting opportunity for reaching tristability for LCP while cancelling it for RCP for a particular band of wavelengths [Fig. 5(b)]. In addition, the bistability field threshold for LCP drops but for RCP it rises [Fig. 5(c)]. Thus, gyrotropy removes the degeneracy in a nonlinear optical response of MPNPs for LCP and RCP driving, which can be used in polarization-sensitive switching discussed in the next section.

B. Temporal dynamics

In this section we study the temporal nonlinear dynamics of the MPNP described by Eq. (9). In Fig. 6, we plot the temporally dependent scattering efficiency for both LCP and RCP waves with a fixed wavelength close to the Fano resonance [see Fig. 5(b)]. The background field starts to grow from zero at t = 0, reaching a saturation level of $E_0 = 50$ statvolt/cm. In a while ($\sim 1 \text{ ps}$) the system comes into the steady state. Then, a Gaussian signal pulse centered at 1.46 ps appears. In a nonmagnetic regime and for g = 0.3 in the case of RCP driving, the system transits from a weak-scattering to a strongscattering steady state with a characteristic switching time ~ 0.5 ps. Remarkably, similar characteristic switching times were obtained for semiconductor microcavities [55,56]. However, for LCP the switching is cancelled and the MPNP comes back to the initial steady state. This happens because for a chosen wavelength of 358 nm the peak signal pulse field overcomes the bistability thresholds for g = 0 and g = 0.3in the case of RCP, but is not large enough to induce a transition for LCP. Although, for the outer plasmon, the steadystate bistability domain is just slightly affected by gyrotropy [Figs. 5(b) and 5(c)], this effect is pronounced enough to induce a difference in dynamical switching.

Finally, we estimate the maximal driving time to avoid particle ablation. To this end, we rely on the results of a previous study on the ablation thresholds for silver particles providing a value of about 3.96 J/cm^2 [57]. Taking into account the amplification of the electric field inside the Ag shell due to plasmonic resonance, we come to the critical illumination

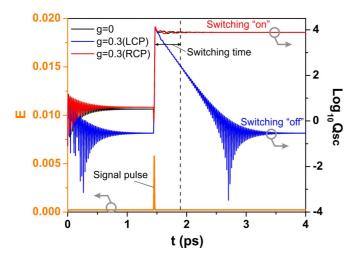


FIG. 6. Temporal dependencies of the scattering efficiency and the dimensionless driving field (orange). The signal pulse provokes a transition from a weak-scattering to a strong-scattering state for g = 0 (black) and g = 0.3 in the case of a right circularly polarized light (red). For a left circularly polarized light, the system returns to the initial state (blue). The parameters are $E_0 = 50$ statvolt/cm (the background field), $E_p = 1265$ statvolt/cm (the pulse peak field), and $\lambda = 358$ nm.

duration after which the MPNP is expected to be burned, \sim 330 ps, which is much longer than the characteristic switching time. Thus, all predicted phenomena are readily observed in experiment.

IV. CONCLUSION AND OUTLOOK

In summary, we have shown that a coated magnetoplasmonic nanosphere with a Kerr-type nonlinear plasmonic shell and a magneto-optic core features optical bistable responses for dipole-type plasmons localized on the outer and inner surfaces of the shell. This bistability can be used to induce switching between the bright outer plasmon and a dark Fano resonance (Fano switching). Once the intensity of the driving light exceeds the critical value, optical tristability arises as a result of overlapping the bistability zones of both eigenmodes. With the help of the dispersion relation method, we have developed a dynamic theory of transitions between nonlinear steady states and estimated the characteristic switching time to be as short as 0.5 ps. The MO effect allowed spectral red and blue tuning of the Fano profile for right and left circularly polarized light, making Fano switching possible for RCP and cancelling it for LCP.

Our results pave the way for using MO elements for tuning the nonlinear dynamical response of nanostructures. For example, nonlinear magnetoactive switchers sensitive to light polarization can significantly increase the number of simultaneously processing channels for on-chip nanoantennas [58]. In analogy to metallic nanoparticles, arrays of MPNPs are expected to support a variety of nonlinear subwavelength dynamical modes in the form of kinks, solitons, oscillons, and spatial patterns [40,59–62], and the MO effect will serve as a powerful tool for steering these modes and searching for novel nonlinear dynamical phenomena.

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