Magneto-optical spaser

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We present an electrodynamical model of a quantum plasmonic device—the magneto-optical (MO) spaser. It is shown that a spherical gain nanoparticle coated with a metallic MO shell can operate as a spaser amplifying circularly polarized surface plasmons. The MO spaser may be used in design of an optical isolator in plasmonic transmission lines as well as in spaser spectrometry of chiral molecules.

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Recently, nanosources of coherent light have attracted significant attention [1]. After [2], different plasmonic-based light sources have been studied theoretically [3–5] and realized experimentally [6–8]. In most cases, such a light source is a pumped optical emitter (a quantum dot or an active molecule) placed inside or near a plasmonic resonator. For the case of a metallic nanoparticle, such a coupled system represents a spaser [2,9–12] or dipole nanolaser [13], whose properties have been intensively studied in detail during the last decade. However, to the best of our knowledge, radiation from all suggested sources is linearly polarized.

Today, interaction of circularly polarized light with matter is becoming an attractive field of research [14–18]. The increasing interest in this topic stems from the fact that sources of circularly polarized light find a number of intriguing applications in molecular sensing [16,17] and quantum information [18]. Sources most commonly used to generate circularly polarized light are either vertical cavity surface-emitting lasers or spin-polarized diodes [15]. The major feature of such sources is that they are diffraction limited.

In this Letter, we suggest a magneto-optical (MO) spaser—a subdiffractive source of circularly polarized light operating in the near field. Employing the description of the gain medium in terms of negative losses, we study its spasing modes. We establish the spaser generation condition and show that for some values of pump intensity, only one of two modes can be realized.

A suggested MO spaser consists of an amplifying core, e.g., a quantum dot or active molecules, coated by a metallic layer exhibiting MO response at optical frequencies [see a schematic drawing in Fig. 1(a)]. The whole core-shell nanoparticle is of subwavelength size. The dielectric permittivity of the gain medium is approximated by the Lorentzian profile with "negative" losses [19,20]. The simplest expression for the permittivity suitable for our purposes may be deduced from the Maxwell–Bloch equations [21]. In the framework of this approach, the evolution of electric field **E** is related to the macroscopic polarization **P** of a gain subsystem via the classical Maxwell's equation, while the dynamics of the polarization and the population inversion of active atoms n are governed by the equations following from the density matrix formalism (see [19] for detailed derivation). The gain atoms embedded into the host medium are modeled as two-level systems with transition dipole moment $\boldsymbol{\mu}$ spread in the host matrix. Assuming harmonic time dependence of the electric field, $e^{-i\omega t}$, we obtain the relation between the polarization P and the electric field E inside the medium, resulting in the following expression for nonlinear permittivity of a gain medium with the anti-Lorentzian profile:

$$\varepsilon_{\text{gain}}(\omega) = \varepsilon_0 + D_0 \frac{\omega_0}{\omega} \frac{-i + \frac{\omega^2-\omega_0^2}{2\omega\Gamma}}{1 + \beta|E(\omega)|^2 + \left(\frac{\omega^2-\omega_0^2}{2\omega\Gamma}\right)^2}, \qquad (1)$$

where $D_0 = 4\pi\mu^2\tau_p n_0/\hbar$ is proportional to the population inversion n_0 in active atoms in absence of plasmonic metal, and, therefore, it describes the pumping rate, $\beta = \mu^2\tau_n\tau_p/\hbar^2$, and $\Gamma = 1/\tau_p$ Here τ_p and τ_n are relaxation times for polarization and inversion, respectively.

The dielectric properties of MO metal, which is assumed to be cobalt, are described with the permittivity tensor having off-diagonal elements accounting for the MO effect:

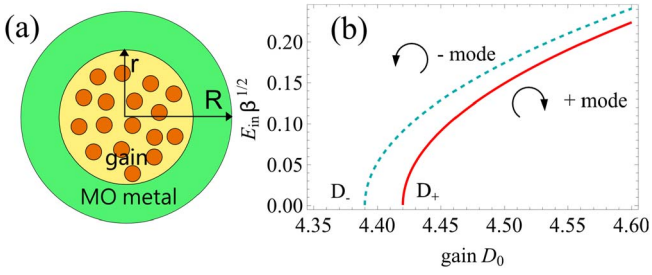

Fig. 1. (a) Schematic drawing of the suggested composite core-shell MO spaser. (b) Amplitudes of spasing modes of the MO spaser versus gain near the bifurcation point.

$$\hat{\varepsilon}_{\mathrm{MO}} = \begin{pmatrix} \varepsilon & ig & 0 \\ -ig & \varepsilon & 0 \\ 0 & 0 & \varepsilon \end{pmatrix}. \quad (2)$$

For values of the diagonal and off-diagonal elements of the permittivity tensor, we use experimental data obtained in [22].

We start our consideration by finding steady-state solutions of an MO spaser with no applied electric field within the quasi-static approximation. As a rule, in the absence of the applied field, there is no field (and no dipole moment) inside the core-shell particle. An exception is the plasmon resonance at which the response of an inclusion to the external field is infinite. For a spherical inclusion, this occurs when $\varepsilon_{\mathrm{incl}} = -2\varepsilon_{\mathrm{host}}$, where $\varepsilon_{\mathrm{incl}}$ and $\varepsilon_{\mathrm{host}}$ are permittivities of the inclusion and the host matrix, respectively. Note that for lossy metal, a gain matrix with $\mathrm{Im}\,\varepsilon_{\mathrm{host}} < 0$ is required [23]. In what follows, we find such a solution with no incident field for an MO spaser with gain core and lossy shell.

Assuming a homogenous nonzero field inside the core, $\mathbf{E}_{\mathrm{core}}(\mathbf{r}) \equiv \mathbf{E}$, the field inside the shell may be sought as $\mathbf{E}_{\mathrm{shell}} = \hat{B}\mathbf{E} - \hat{C}\mathbf{E}/\rho^3 + 3(\hat{C}\mathbf{E}, \mathbf{n})\mathbf{n}/\rho^3$ and the field outside nanoparticle as $\mathbf{E}_{\mathrm{ext}} = -\hat{A}\mathbf{E}/\rho^3 + 3(\hat{A}\mathbf{E}, \mathbf{n})\mathbf{n}/\rho^3$, where $\hat{A}$, $\hat{B}$, and $\hat{C}$ are some unknown antisymmetric tensors, and ρ is the distance from the center of the particle. Employing the usual boundary conditions, we can find the unknown tensors $\hat{A}$, $\hat{B}$, and $\hat{C}$ and write the condition of the existence of a nonzero internal field inside the nanoparticle at zero incident field in the form of an eigenvalue problem (see [24]):

$$\varepsilon_{\mathrm{gain}}\mathbf{E} = [\varepsilon(\hat{B} + 2\hat{C}/r^3) + \hat{G}(\hat{B} - \hat{C}/r^3)]\mathbf{E} = \hat{M}\mathbf{E}, \quad (3)$$

where $\hat{G} = (\hat{\varepsilon}_{\mathrm{MO}} - \hat{\varepsilon}_{\mathrm{MO}}^T)/2$ stands for the nondiagonal part of the permittivity tensor of the MO shell. Due to the dependence of $\varepsilon_{\mathrm{gain}}$ on E^2 and pumping D_0, this equation can be treated as the condition for spasing. We only consider solutions with $E_z = 0$, for which the polarization is perpendicular to the magnetization vector of the MO shell. From the symmetry of the problem, it follows that the tensor $\hat{M}$ has the form

$$\hat{M} = \begin{pmatrix} M_{1,1} & M_{1,2} \\ -M_{1,2} & M_{1,1} \end{pmatrix}, \quad (4)$$

and, therefore, its two eigenvalues are given by $\lambda_\pm = M_{1,1} \pm iM_{1,2}$ with two eigenvectors corresponding to the right and left circular polarizations of the electric field inside the amplifying core. Thus, the condition for spasing (3) can be recast as

$$\varepsilon_{\mathrm{gain}}(\omega, E_{\mathrm{in}}, D_0) = \lambda_\pm, \quad (5)$$

$$\lambda_\pm = \frac{(-2 + 2\varepsilon \mp g)(\varepsilon \pm g)r^3 - (2 + \varepsilon \pm g)(2\varepsilon \mp g)R^3}{(-2 + 2\varepsilon \mp g)r^3 + (2 + \varepsilon \pm g)R^3}. \quad (6)$$

Equation (5) defines two spasing modes with circular polarizations. As we have shown for the nongirotropic spaser [24], their dependence on gain exhibits the Hopf bifurcation. Indeed, the right-hand side of Eq. (5) does not depend on E_{in} and D_0 and hence has a nonvanishing imaginary part for all frequencies in case of lossy metal. At the same time, $\varepsilon_{\mathrm{gain}}$ has an arbitrary small imaginary part for vanishing D_0. Threshold gain for spasing D_{th}, which is the smallest possible gain D_0 for which Eq. (5) is satisfied, is achieved for $E_{\mathrm{in}} = 0$.

To model the gain medium, we use values of parameters that are typical for organic dyes [25]: $\omega_0 = 2$ eV, $\varepsilon_0 = 2$, and $\gamma = 0.05$ eV. The electric field is measured in the units of $\beta^{-1/2}$, so its value is of no importance. The radii of the gain core and MO shell are $r = 20$ nm and $R = 1.1r$.

Our calculations show that the two spasing modes have different spasing frequencies $\omega_\pm$ and different spasing thresholds $D_\pm$. In Fig. 1(b) we depict the mode amplitudes (the field inside the amplifying core E_{in}) versus gain D_0 for an MO spaser. One can see that in the interval of gain values $4 < D_0 < 5$ the two spasing modes arise with square-root-like dependence of amplitudes on gain. The calculated spasing frequencies are $\omega_- = 1.976$ eV and $\omega_+ = 1.977$ eV.

Figure 1(b) shows that there exists a range of gain D_0 for which only a left or right circularly polarized spasing mode is emitted. Which particular mode is realized depends on the direction of the magnetization vector. The difference between the thresholds of the modes is simply the consequence of the circular dichroism of the MO shell. Indeed, in the circular polarization basis the permittivity tensor of MO metal is $\hat{\varepsilon}_{\mathrm{MO}} = \mathrm{diag}[\varepsilon + g, \varepsilon - g, \varepsilon]$, so that $\varepsilon + g$ eigenvalue corresponds to $\mathbf{E}_- = (1, -i, 0)^T$ polarization and $\varepsilon - g$ to $\mathbf{E}_+ = (1, i, 0)^T$, respectively. This means that the effective permittivity of MO metal differs for right and left polarized modes, leading to different Q factors for these modes. As a result, the threshold for one of the two modes is smaller.

The behavior of the MO spaser above the highest threshold D_+ is strongly affected by the structure of dipole transitions of the gain medium [26]. If the transition dipole moments between the excited and ground states of the dye molecule are linearly polarized, both modes obtain the energy from the same pumping D_0 [see Eq. (1)], and it is expected that due to the mode competition mechanism only the mode with the lower threshold is realized. If the transition dipole moments are circularly polarized, then the pumping rates for each mode are different and one can expect coexistence of the two nonzero modes with different frequencies. In this case, beatings of the total dipole moments with the beating frequency proportional to the difference $\omega_+ - \omega_-$ may occur. Note that the present model cannot reveal above-threshold behavior and a more detailed quantum-mechanical approach is needed.

For the core-shell spaser considered above, the developed approach predicts the value of threshold gain as $D_{\mathrm{th}} \simeq 4.5$, leading to the bulk gain coefficient $k'' \simeq D_{\mathrm{th}}\omega/(2c) \simeq 10^5$ cm^{-1}. This value is unachievable for typical organic dyes [25] even for full population inversion. Fortunately, the described effect can be observed in systems using plasmonic metals and dielectric low-loss MO materials, such as yittrium–iron–garnet (YIG).

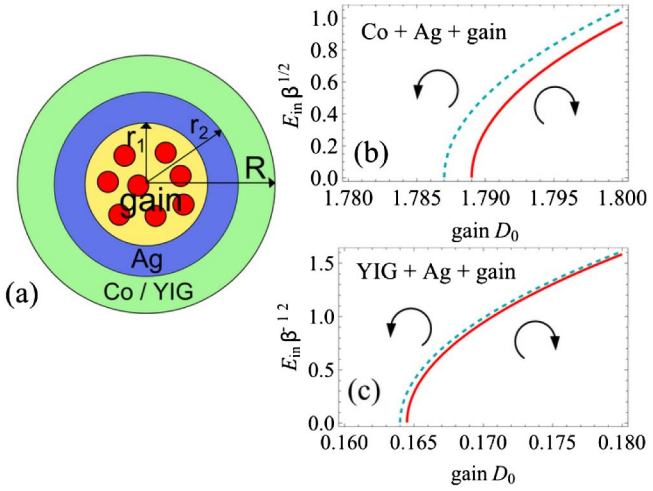

Fig. 2. (a) Alternative design of the MO spaser allowing for low-threshold spasing. (b), (c) Amplitudes of spasing modes versus gain for Co + Ag and YIG + Ag spasers, respectively.

To overcome large ohmic damping and unrealistic gain required for operation of the Co-based spaser, we suggest two modifications of the original design: a spherical gain core coated first with a silver shell and on top of it with another shell of MO cobalt or YIG [Fig. 2(a)]. Due to the presence of the silver shell, such a structure forms a high-Q plasmonic resonator exhibiting MO properties. The amplitudes of the two circularly polarized modes are governed by the same Eq. (5) in which modified $\lambda_{\pm}$ are now defined by boundary conditions at three interfaces $r_1 < r_2 < R$.

The threshold gain of these structures depends on the radii of the gain core and the silver and MO shells and the background permittivity of gain medium ε_0. Gain required for spasing can be minimized by choosing optimal parameters for both structures. In addition, one should keep in mind that the plasmon resonance of core/shell nanoparticles has to be tuned to the frequency range of the MO resonance of MO material.

Calculations show that the threshold for the Co/Ag spaser can be as low as $D_{\text{th}} \approx 1.8$ for $r_1 = 0.6R$, $r_2 = 0.95R$, and $\varepsilon_0 = 5$. In order to approximate permittivity for silver, we use experimental data from [27]. When the thickness of the cobalt layer is larger than $0.05R$, the spasing threshold quickly increases almost to its original unrealistic value $D_{\text{th}} \approx 3 \div 4$. For the case of a YIG/Ag spaser, the conditions for spasing are even more favorable: by choosing $r_1 = 0.7R$, $r_2 = 0.9R$, and $\varepsilon_0 = 2$, we attain a decrease of the threshold up to $D_{\text{th}} = 0.17$, leading to a bulk gain coefficient of approximately $k'' \simeq 3 \times 10^3$ cm^{-1}, which is achievable for organic amplifying media.

In conclusion, we have suggested an MO spaser representing an ultrasmall source of coherent circularly polarized light. Unlike a nonmagnetic spaser, this light source has two spasing modes with left and right circular polarizations of the dipole moments. Each of the modes has a different pumping threshold. Due to the competition of modes, only the mode with the lower pumping threshold survives. As a result, a specific circular polarization is realized. This is especially important for prospective applications in quantum information, where manipulations of qubits require illumination with circularly polarized light.

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