Gain-Assisted Surface Plasmon Polaritons: Time Domain Analysis with Experimentally Fitted Organic Dye Models

Shaimaa Azzam[1], Nikita Arnold[2], Ludmila Prokopeva[3], Zhaxylyk Kudyshev[1], and Alexander V. Kildishev[1,*]
[1]School of Electrical and Computer Engineering and Birck Nanotechnology Center, Purdue University, West Lafayette, IN 47907, USA
[2]Institute of Applied Physics, Johannes Kepler University, Altenberger Straße 69, 4040, Linz, Austria
[3]Novosibirsk State University, Pirogova 2, 630090, Novosibirsk, Russia
*Author e-mail address: kildishev@purdue.edu**

Abstract: Kinetic parameters of a six-level model are obtained by matching the experimental data for a fluorescent dye to study loss-compensation in the surface plasmon-polariton propagating along a silver film covered with a gain layer.

OCIS codes: (240.6680) Surface plasmons; (250.5403) Plasmonics; (240.6690) Surface waves;

1. Introduction

One major limitation in plasmonic-based systems is the large damping due to losses in metals. Overcoming these losses has been an active research subject [1, 2]. A promising solution is incorporating optical gain material into the dielectric part of a nanoplasmonic system [2]. Design and optimization of nanolasers and spasers [3-5] is yet another application area, where combining optical gain inclusions with plasmonic elements becomes imperative.

Among the wide variety of optical gain materials, organic dyes are known for their broad spectra covering normally up to several hundreds of nanometers making them attractive for wavelength tunability. The development and optimization of active nanoplasmonic devices should be supported and facilitated by accurate theoretical models and numerical techniques that can adequately describe the interaction between light and organic dyes. Accurate numerical simulations of lasing dynamics of organic dyes in time-domain are currently built on a semi-classical model. In the model the atomic system is treated quantum mechanically and electromagnetic wave is treated classically. These gain material models relying on single- and two-electron four-level rate equations for electron populations have proven to be efficient in handling a wide range of gain materials [6,7]. However, some organic dyes can exhibit non-negligible split transitions requiring more advanced and accurate material models.

In this work, a six-level single-electron system is used to fit the parameters of Thermo-Scientific Green fluorescent dye in a polystyrene film obtained from a pump-probe experiment. The six-level kinetic model takes into account split transitions, which play an increasingly significant role in more complex gain materials allowing lasing to occur at multiple wavelengths. The optical gain introduced by the “Firefli* Fluorescent Green” dye (Thermo-Scientific, Waltham, MA, USA) is then used to compensate for surface plasmon polariton (SPP) losses in a Kretschmann-Raether configuration, Fig. 2, similar to that reported experimentally in [2]. Enhancement of the reflectivity signal is observed to be as high as 525% for concentration of the dye molecules of 2×10^{20} cm^{-3}.

2. Analysis and results

Light-matter interaction in optical gain media is modeled using a semi-classical model in which atom is treated quantum-mechanically and electromagnetic wave is treated classically. The atom in our analysis is modeled by a six-level single-electron system allowing electronic transitions between the levels shown in Fig. 1. The system is described by a set of rate equations that quantify the transitions between different sublevels. Rate equations are then coupled to the standard FDTD method [6] via auxiliary differential equations (ADE) [8]. For the Thermo-Scientific Green fluorescent dye, the wavelengths corresponding to energy differences between different atomic levels shown in Fig. 1 are as follow: λ_{50} = 438 nm, λ_{40} = 466 nm, λ_{32} = 513 nm, and λ_{31} = 486 nm. The dye is pumped at 438 nm from the backside of the prism while the probe is incident through a glass prism as in Fig.2. The pulse width for the pump signal is 1 ps and the probe signal is 10 fs. The probe signal is delayed by 5 ps from the pump. In our Kretschmann-Raether configuration with a 50-nm silver film, the SPP resonance condition at 513 nm corresponds to an incidence angle of 46.5º which is represented by the dip in the reflectivity curve as shown in Fig. 3. Dielectric functions of the silica prism and the silver film are taken form Palik [9]. A 2-D FDTD analysis is carried out for the experimental setup (Fig. 2) with and without the pumping signal. The results are shown in Fig. 3: the solid black line represents reflectivity without pumping (no gain), the dashed red line corresponds to reflectivity in the presence of gain with dye molecule density $N_d = 2\times10^{19}$ cm^{-3} and the solid blue line indicates the presence of gain with dye molecule density $N_d = 2\times10^{20}$

cm^{-3}. Relative enhancement in reflectivity is 180% with a concentration of dye molecules of 2×10^{19} cm^{-3} and around 525% with concentration of dye molecules of 2×10^{20} cm^{-3}. These results suggest that a complete suppression of the SPP losses is achievable in our study with correct concentration of dye molecules.

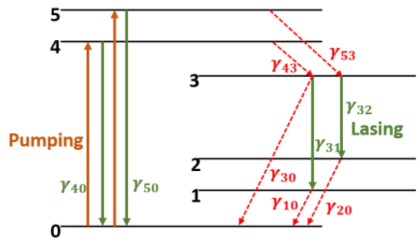

Fig. 1. Jablonski diagram of the six-level atomic system with stimulated transitions and relaxations.

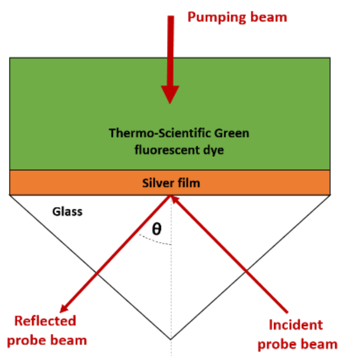

Fig. 2. Schematic of SPP excitation in the Kretschmann-Raether configuration.

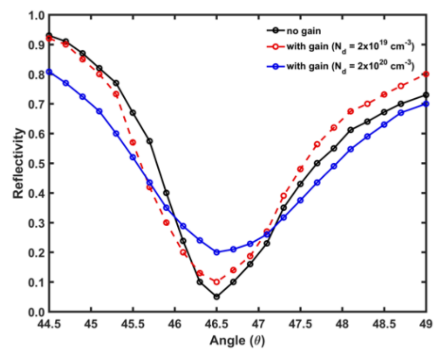

Fig. 3. Reflectivity with respect to incident angle (θ) with and without gain at different concentrations of dye molecules.

3. Summary

A six-level single-election kinetic model (in which additional split transitions are fully accounted for) is employed to simulate the stimulated emission of organic dye molecules. Time domain modeling of the gain-assisted SPP adjacent to a polystyrene layer mixed with the "Firefli* Fluorescent Green" dye (Thermo-Scientific, Waltham, MA, USA) has been performed. Up to 525% and 180% enhancement in reflectivity is observed for dye concentrations of 2×10^{20} and 2×10^{19} cm^{-3} respectively. Our simulations are in good agreement with the published experimental results considering SPP propagation with amplification in a similar geometry [2], where 280% enhancement in reflectivity was observed with a different dye (R6G) in the PMMA film for a dye concentration of 1.26×10^{19} cm^{-3}. The proposed modeling approach stimulates further experimentally fitted numerical studies of quantum emitters coupled with plasmonic nanostructures, such as spasers, plasmonic nanolasers, and plasmonic waveguide amplifiers.

The authors acknowledge ARO grant W911NF-13-1-0226 and AFOSR MURI grant FA9550-12-1-0389.

4. References

[1] S. Xiao, V. P. Drachev, A. V. Kildishev, X. Ni, U. K. Chettiar, H.-K. Yuan, and V. M. Shalaev, "Loss-free and active optical negative-index metamaterials," Nature **466**, 735-738 (2010).
[2] M. A. Noginov, V.A. Podolskiy, G. Zhu, M. Mayy, M., Bahoura, J.A. Adegoke, B.A.Ritzo, and K. Reynolds, "Compensation of loss in propagating surface plasmon polariton by gain in adjacent dielectric medium", Optics Express **16,** 1385-1392 (2008).
[3] D. J. Bergman and M. I. Stockman, "Surface plasmon amplification by stimulated emission of radiation: quantum generation of coherent surface plasmons in nanosystems," Phys. Rev. Lett. 90, 027402 (2003).
[4] X. G. Meng, A. V. Kildishev, K. Fujita, K. Tanaka and V. M. Shalaev, "Wavelength-tunable spasing in the visible," Nano. Lett. 13, 4106-4112 (2013).
[5] N. Arnold, C. Hrelescu, and T. A. Klar, "Minimal spaser threshold within electrodynamic framework: Shape, size and modes," Annalen der Physik **528**, 295-306 (2016).
[6] A. Taflove and S. Hagness, " Computational Electrodynamics: The Finite-Difference Time-Domain Method," Artech House, 2005, 3rd ed.
[7] L. J. Prokopeva, J.Trieschmann, T.A. Klar, and A.V. Kildishev, "Numerical modeling of active plasmonic metamaterials," *SPIE Optical Systems Design*, pp. 81720B-81720B. International Society for Optics and Photonics, (2011).
[8] S. V. Zhukovsky and D. N. Chigrin, "Numerical modelling of lasing in microstructures," Phys. Status Solidi (B) vol. **244**, 3515-3527 (2007).
[9] Palik, Edward D. *Handbook of optical constants of solids*. Vol. 3. Academic press, (1998).