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# Hybridization influence on the plasmon-mediated lasing effect in active metamaterials

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# ABSTRACT

The plasmon-mediated lasing with orders of magnitude amplification is available for a double-fishnet structure, however, it gradually disappears for multilayered fishnet stacks because of the layer-to-layer plasmon hybridization, while instead a broadened incoherent transmission with full loss compensation can always be obtained regardless of the stacking layers.

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## 1. Introduction

Electromagnetic metamaterials with subwavelength structures are very attractive artificial materials for the variety of novel properties which are not found in nature, such as negative refraction [1], super imaging [2], cloaking [3], optical activity [4], surface plasmon amplification by stimulated emission of radiation (spaser) [5-7], and the plasmonic analogy of electromagnetically induced transparency [8,9]. However, metallic loss poses a great challenge to get various responses in metamaterials applicable. As a readily conceivable method, active medium can be introduced to compensate the tremendous losses confronted in nearly all the uncommon properties proposed in metallic metamaterials. Recently, the socalled lasing spaser (or spasing), originally proposed by Zheludev et al. [6], is of great interest which can realize a great transmission amplification in the composite metamaterial of a subwavelength metallic structure combined with an active medium (for example, quantum dots) and thus contributes to the exploration of nanolaser [10,11]. According to literatures, for a lasing spaser process, the excited state of an active medium, pumped by an external control power, does not radiate to outer environment, but it transfers the energy losslessly to the resonant state of the surface plasmons in the adjacent metallic structure by producing excitons coherent to the incident light (so-called stimulated emission of radiation), and hence a lasing spaser was considered as a new type of nanolasing source, which differs from an ordinary laser in several aspects. Specially speaking, a lasing spaser takes the advantage of the resonant metallic nano-system to replace a conventional laser cavity, and it amplifies the surface plasmons instead of photons as do in conventional lasers. In particular, it can reach a large amplification with a relatively small gain coefficient as compared with other common gain-assisted mechanisms.

To obtain the maximum magnitude amplification of the spasing transmission, the underlying resonant mode from the metallic structure is a very important factor. For example, the asymmetrically split ring (or arc pair) can produce a coherent resonance (or trapped mode) with high quality factor which contributes to the spasing efficiency. N. Papasimakis et al. studied the coherent and incoherent resonances in ordered and disordered arrays; they distinguished that a coherent response exhibiting a collective resonance would disappear if an originally ordered metamaterial is rearranged in disorder, while an incoherent resonance does not show obvious sensitivity to the periodic arrangement [12]. In our previous work, we used a Fabry–Perot cavity slab with Lorentz dispersion in the index of refraction to model the dependence of giant magnitude amplification on the gain coefficient [13].

However, all the spasing metamaterials published to date, including the above-mentioned works [6,10,13], are generally in two

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**Fig. 1.** (Color online.) The schematic illustration of the bulk metallic fishnet metamaterial with active layer as the spacer. (a) Bulk fishnet metamaterial with four-layer metallic fishnet structures, the yellow area indicates an *xy*-face of the selected unit cell. (b) The unit scales for the fishnet metamaterial.

dimensions, that is, in planar configurations of metallic structures. To the best knowledge of ours, no papers have been presented about the layer-to-layer influence on the spasing efficiency when planar metamaterials are stacked into multilayer ones. In this work, we investigate the transition of the spasing transmission in a fishnet metamaterial by increasing the stacking layers. The simulation results show that sharp spasing peak with orders of magnitude amplification would gradually degenerate to a level around 0 dB, for which the reason is attributed to the layer-tolayer coupling effect (i.e., plasmon hybridization).

#### 2. Numerical model

According to the work of Zheludev et al., the active medium, such as a semiconductor pumped by optical or electrical signal, can be characterized by a gain coefficient  $\alpha$  with the value of  $(2\pi/\lambda) \operatorname{Im}(\sqrt{\varepsilon' + i\varepsilon''})$ , where  $\varepsilon'$  and  $\varepsilon''$  are the real and imaginary parts of the electric permittivity of the active medium, respectively [6]. In this work, this kind of active medium is used as the spacer of the metallic multilayers composed of fishnet structure. Fig. 1(a) shows schematically the bulk fishnet metamaterial with four layers of fishnets, where the geometric parameters in Fig. 1(b) are as follows:  $a_x = 160$  nm,  $a_y = 100$  nm, and  $L_x = L_y = 300$  nm. The 20-nm-thick metal in fishnet shape is silver with Drude-type dispersion ( $\omega_p = 1.37 \times 10^{16} \text{ s}^{-1}$  and  $\gamma = 8.5 \times 10^{13} \text{ s}^{-1}$ , see Refs. [14] and [15]). In addition, the layer thickness of the active dielectric spacer is 30 nm (i.e., the gap between the adjacent metallic layers) and its real part of permittivity is 2.5. In our numerical simulations based on the full-wave finite element method, the polarized incident wave with electric field in the x direction and magnetic field in the y direction is satisfied by applying the perfect electric and magnetic boundary conditions, respectively [15,16]. For clarity, the unit cell in Fig. 1(b) is referred to as a double-layer fishnet structure, which is the primary functional stack of a fishnet metamaterial since no monolayer metallic fishnet is expected to exhibit the same resonant mode. On the analogy of this, the illustration in Fig. 1(a) is a four-layer bulk fishnet metamaterial, and so on.

# 3. Numerical results

What characterizes the mechanism of stimulated emission of radiation is the amplification at the resonance, but no obvious enhancement could be found away from the resonance regime. Fig. 2(a) shows the transmission results of a double fishnet layers assisted by the active spacer, from which the stimulated emission of the radiation by the fishnet metamaterial can be confirmed in a similar manner with the result of the ASR metamaterial. The maximum enhancement of magnitude reaches 46.8 dB at 580 THz for  $\alpha = 1.09 \times 10^4$  cm<sup>-1</sup>, almost 220 times of the incident inten-



**Fig. 2.** (Color online.) (a) The lasing effect by stimulated emission of left-handed radiation in the double fishnet metamaterial for various gain coefficients. The opposite current distributions on the inner surfaces of the double metallic layers shown in (b) and (c) confirm the left-handed resonant mode.



Fig. 3. Transition of the left-handed transmission with stacked metallic layers at the lasing gain coefficient  $(1.09 \times 10^4 \text{ cm}^{-1})$  of the double-layer fishnet structure in Fig. 2.

sity, while for a spacer without gain/attenuation ( $\alpha = 0 \text{ cm}^{-1}$ ), the same resonance is completely lost because of the severe loss (see the black data line). A detailed dependence of the resonance amplification on the gain coefficient can be found in Refs. [13] and [16]. It is worthy to notice that, as an analogy to the leakage of radiation through the output coupler of a laser cavity, a deliberate small asymmetry in the ASR structure is introduced to leave a fraction of the accumulated energy of plasmonic oscillation to be radiated, turning the otherwise dark mode into a bright one [6]. Here, for the resonant behavior in this fishnet structure, the induced current distributions at 580 THz for  $\alpha = 1.09 \times 10^4$  cm<sup>-1</sup> in Figs. 2(b) and 2(c) identify that it is a resonant mode same to the left-handed response in fishnet metamaterials [6]. Therefore, the resonance mode itself is radiative, and thus no additional asymmetry in the fishnet structure is necessary for its plasmon-mediated lasing.

Although the double-layer fishnet structure exhibits an intriguing spasing effect with orders of magnitude enhancement for the resonant transmission peak, it remains an interesting work, yet to be investigated, what will happen for the multilayer metamaterials as far as the plasmon-mediated lasing property is concerned. In Fig. 3, the simulation results reveal that, with the increasing of the stacking layers, the spasing spectrum becomes broader while the resonance enhancement gets weaker in magnitude. That is to say, the giant magnitude amplification for a spasing characteristic prefers a planar structure. To explain this, the concept of "coherent metamaterial" should be of great help. According to Refs. [6] and [12], the spasing effect could be obtained provided that a single mode with high quality factor (such as a trapped mode) happens, this is true for an elementarily functional fishnet metamaterial (double-layer structure). However, for the multilayer structures, the layer-to-layer strong coupling leads to a hybridization effect that will split the single resonant mode  $\omega_0$  in double-layer fishnet into two or more resonant modes while increasing the stacking layers [Fig. 4(a)]. This can be confirmed from the multiple resonant peaks shown in Figs. 3(b) and 3(c). According to the schematic of plasmon hybridization effect in Fig. 4, N coupled layers will result in N-1 resonant modes, unless some of the hybridized resonant modes are degenerated to some extent. Eventually, a broad band around frequency  $\omega_0$  will be formed for a numerously stacked fishnet metamaterial [Fig. 4(a)]. As a consequence, plasmon hybridization [4,16–20] leads to the excitation of coupled modes with incoherent characteristic as well as low Q factor. Note that the coherent resonance with narrow resonant spectrum in a planar metamaterial with ordered arrangement could also be eliminated in disordered arrays of the meta-structure, as reported recently by N. Papasimakis et al. [12].

On the other hand, it is found in Fig. 3(d) that the magnitude of the originally spasing behavior will decrease to a level less than 0 dB for the 50-layer fishnet metamaterial at  $\alpha = 1.09 \times 10^4$  cm<sup>-1</sup>, which indicates an incomplete loss compensation. Therefore, it raises the question whether complete loss compensation is available in multilayer optical metamaterials, especially for large stacking numbers in the electromagnetic propagation direction. To demonstrate this question, a 100-layer fishnet



**Fig. 4.** (Color online.) (a) Split resonance frequencies for the plasmon hybridization in layer-to-layer coupling fishnet metamaterials. (b) Three hybridization modes for a four-layer fishnet metamaterial. Arrows denote the induced surface current, while "+" and "-" present the locations of charge accumulation.

metamaterial, together with a 50-layer one, was simulated to investigate the magnitude enhancement under the assistance of an optically active medium. Fig. 5(a) shows the dependence of the maximum resonant transmission on gain coefficient, it is clear that a complete loss compensation, corresponding to approximately 0 dB maximum  $S_{21}$ , is available for a value of gain coefficient around  $9.0 \times 10^3$  cm<sup>-1</sup>, and no amplified transmission magnitude above 0 dB is found in our simulations for a sufficiently-thick fishnet metamaterial no matter what value for the gain coefficient will be assumed. It seems counterintuitive for the maximum transmission to be reduced when the gain coefficient is increased more than a threshold value around  $9.0 \times 10^3$  cm<sup>-1</sup>. To the best knowledge of ours, a systematic analysis is yet to be explored on this issue since it remains unclear except for some simply explanations in Refs. [6] and [13]. Nevertheless, as far as this simulation result is concerned, it is consistent with the literature (e.g. Ref. [6]).

In addition, the broadened, lossless resonance transmission at  $\alpha = 9.0 \times 10^3$  cm<sup>-1</sup> can be found from the transmission spectrum for the 100-layer fishnet case [Fig. 5(b)], and correspondingly Fig. 5(c) presents the local magnetic field amplitude along the centers of the individual active layers (in the propagation direction), from which the lossless propagation could be visually evaluated. Though the electromagnetic waves can propagate through the multilayered structure without attenuation, we also notice that there are obvious amplitude variations of the magnetic field from layer to layer. Such variations should be attributed to the numerical inaccuracy since these field amplitudes are obtained in the middle positions of every active layer, where strong resonant field localizations will beat the simulation accuracy. In fact, we also simulated bulk optical metamaterials with stacks more than 100 layers, the loss compensation is expectably no difference for the critical gain coefficient about  $9.0 \times 10^3$  cm<sup>-1</sup>, since the balance between the metallic loss and active amplification is established for the critical gain coefficient case and the plasmon hybridization effect between layers has been counted.

## 4. Summary and conclusion

We have investigated the optically active fishnet metamaterial at the resonance frequency near 580 THz (517 nm wavelength), from which different enhancement responses of the electromagnetic transmission are found between the double-layer and multilayer fishnet metamaterials. The plasmon-mediated lasing is available only for a double-layer fishnet structure, about 220 times of the maximum magnitude enhancement of the transmission at the gain level of  $\alpha = 1.09 \times 10^4$  cm<sup>-1</sup>. In contrast, it is indicated that for multilayer fishnet metamaterials the sharp lasing peak of the double-layer fishnet structure is readily transformed to an incoherent broadened transmission with less magnitude amplification



**Fig. 5.** (Color online.) Loss compensated propagation in optically active bulk fishnet metamaterials. (a) Dependence of the maximum left-handed transmission on gain coefficient, with black squares for 50-layer fishnet metamaterial, and red circles for 100-layer fishnet metamaterial. (b) and (c) show the transmission spectrum and amplitude of local magnetic field along the centers of the propagation layers, respectively, for the 100-layer fishnet metamaterial at a gain coefficient of  $9.0 \times 10^3$  cm<sup>-1</sup>.

because of plasmon coupling/hybridization between the neighboring layers. The results show that the coherent transmission amplification of the stimulated emission of radiation will be disturbed when stacking layers of the multilayer metamaterial increase. Nevertheless, complete loss compensation is always available for the multilayer cases.

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