

CHCRUS

This is the accepted manuscript made available via CHORUS. The article has been published as:

Stockman Replies:

Mark I. Stockman Phys. Rev. Lett. **107**, 259704 — Published 13 December 2011 DOI: 10.1103/PhysRevLett.107.259704

Stockman Responds to Comment by Pendry and Maier

Mark I. Stockman¹

¹ Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30303, USA

In their Comment [1] Pendry and Maier (PM) disagree with the conclusion of our Letter [2] that full loss compensation in a dense resonant plasmonic gain metamaterial brings about spasing, [3] clamps the population inversion, and eliminates the net gain. PM's assertion is based on their understanding that "Stockman considered a single cell..." Below we show this statement and conclusions of PM are wrong. Our Letter [2] is correct and stands as originally formulated.

In reality, citing [2]: "Consider a small piece of this metamaterial with sizes much greater that the unit cell but much smaller than λ , which is a metamaterial itself.". Theory of spasing is applied to this metamaterial as a whole, as has always been done, not to the unit cell. Therefore, it completely takes into account the interactions between cells of the system.

The invalidity of the contention of PM that we apply this eigenmode equation only to the unit cell is already obvious from the fact that localization/delocalization of plasmonic eigenmodes has been a subject of our extensive research. Our Ref. 4 confirmed that there are eigenmodes of all localization radii, from size of the unit cell up to the size of the entire system, which coexist at close eigenvalues (frequencies). Note that this phenomenon known as inhomogeneous localization (delocalization) has been predicted for the quasistatic eigenmodes [4–6] and later confirmed experimentally. [7, 8] This can be illustrated by Fig. (2) of Ref. 4 where the eigenmodes with the eigenvalues $s_n = 0.2$ and $s_n = 0.2011$ are clearly delocalized over the entire system containing over $\sim 10^3$ of the units cells (monomers). In theory of spaser, see Fig. (2) (b)-(c) of Ref. 9, the spasing modes are delocalized over all the six nanospheres constituting the spaser.

The entire Comment [1] is based on the disputed premise that Ref. 2 considers spasing in a single unit cell. There is also another mistake in this Comment to be addressed. This is the statement [1] that for spasing one should take into account "the dominant decay channel namely tunneling into adjacent cells through overlap of the resonant fields", and that "neighboring cells will have an effective capture rate $i\Sigma(\mathbf{r})...$ " This imaginary amplitude is considered as an additional loss for spasing but not for amplification. This idea of PM is in conflict with fundamental principles of physics.

In periodic metamaterials, as in any crystals, the overlap of the unit-cell wave functions causes *coherent* coupling between them and not any relaxation or scattering. This is a general tenet of physics established by the classic work of Felix Bloch [10]. The Comment [1] is incompatible with this principle. In reality, the transfer amplitude between the unit cells is real, leading to the formation of energy bands and not to any relaxation, and it will not hamper spasing. In fact, the SP eigenproblem Eq. (1) of Ref. 4 are real and independent from the permitivitties, which, including their imaginary parts, are fully taken into account in our theory through the corresponding Green's functions to enter correctly the results – Eqs. (5)-(13) of Ref. 2.

The well-studied distributed feedback (DFB) lasers [11] are, in fact, periodic systems (gratings) with gain. Deliberately, the DFB gratings are made very weak, so the coupling between the unit cells is very large. PM's theory[1] would imply that the DFB lasers would not generate but they do. Similarly, periodic plasmonic meta-materials with gain are DFB spasers, which, as any DFB lasers, will generate at band-edge modes.

As follows from our results, [2, 3, 12, 13] the conditions for spasing are realistic for existing semiconductor or laser-dye gain media. Therefore, it is not surprising that a significant number of generating spasers have been reported in experiments, [14–18] and more are in progress.

In metamaterials, losses is a major problem severely hampering fundamental effects and meaningful applications. Despite a large effort to compensate loss by gain, there is a little progress. In the literature, there is only one successful compensation of losses claimed [19]. (Note, that in Ref. 19 the 280 nm unit cell is not deeply subwavelength, and that system is a nanolayer rather than a 3d metamaterial.) There have also been multiple attempts to compensate loss by gain for surface polaritons [20–23]. None achieved the full loss compensation, quite possibly, due to unintended spasing on defect-localized modes. Thus this Comment [1] is "at odds" with a large number of experiments.

This work was supported by Grant No. DEFG02-01ER15213 from the Chemical Sciences, Biosciences and Geosciences Division and by Grant No. DE-FG02-11ER46789 from the Materials Sciences and Engineering Division of the Office of the Basic Energy Sciences, Office of Science, U.S. Department of Energy, and by a grant from the U.S.Israel Binational Science Foundation.

- [1] J. B. Pendry and S. A. Maier, Phys. Rev. Lett. (2011).
- [2] M. I. Stockman, Phys. Rev. Lett. 106, 156802 (2011).
- [3] D. J. Bergman and M. I. Stockman, Phys. Rev. Lett. 90, 027402 (2003).

- [4] M. I. Stockman, S. V. Faleev, and D. J. Bergman, Phys. Rev. Lett. 87, 167401 (2001).
- [5] M. I. Stockman, L. N. Pandey, and T. F. George, Phys. Rev. B 53, 2183 (1996).
- [6] M. I. Stockman, Phys. Rev. E 56, 6494 (1997).
- [7] K. Seal, A. K. Sarychev, H. Noh, D. A. Genov, A. Yamilov, V. M. Shalaev, Z. C. Ying, and H. Cao, Phys. Rev. Lett. **94**, 226101 (2005).
- [8] V. Krachmalnicoff, E. Castanie, Y. D. Wilde, and R. Carminati, Phys. Rev. Lett. (2010).
- [9] K. Li, X. Li, M. I. Stockman, and D. J. Bergman, Phys. Rev. B 71, 115409 (2005).
- [10] F. Bloch, Z. Phys. A 52, 555 (1929).
- [11] H. Ghafouri-Shiraz, Distributed Feedback Laser Diodes and Optical Tunable Filters (J. Wiley, West Sussex, England; Hoboken, NJ, 2003).
- [12] M. I. Stockman, Journal of Optics **12**, 024004 (2010).
- [13] M. I. Stockman, Phil. Trans. R. Soc. A 369, 3510 (2011).
- [14] M. T. Hill, M. Marell, E. S. P. Leong, B. Smalbrugge, Y. Zhu, M. Sun, P. J. van Veldhoven, E. J. Geluk, F. Karouta, Y.-S. Oei, et al., Opt. Express 17, 11107 (2009).
- [15] M. A. Noginov, G. Zhu, A. M. Belgrave, R. Bakker, V. M. Shalaev, E. E. Narimanov, S. Stout, E. Herz, T. Sutee-

wong, and U. Wiesner, Nature 460, 1110 (2009).

- [16] R. F. Oulton, V. J. Sorger, T. Zentgraf, R.-M. Ma, C. Gladden, L. Dai, G. Bartal, and X. Zhang, Nature 461, 629 (2009).
- [17] R.-M. Ma, R. F. Oulton, V. J. Sorger, G. Bartal, and X. Zhang, Nat. Mater. 10, 110 (2010).
- [18] R. A. Flynn, C. S. Kim, I. Vurgaftman, M. Kim, J. R. Meyer, A. J. Mkinen, K. Bussmann, L. Cheng, F. S. Choa, and J. P. Long, Opt. Express 19, 8954 (2011).
- [19] S. Xiao, V. P. Drachev, A. V. Kildishev, X. Ni, U. K. Chettiar, H.-K. Yuan, and V. M. Shalaev, Nature 466, 735 (2010).
- [20] M. A. Noginov, G. Zhu, M. Mayy, B. A. Ritzo, N. Noginova, and V. A. Podolskiy, Phys. Rev. Lett. **101**, 226806 (2008).
- [21] J. Grandidier, G. r. C. des Francs, S. b. Massenot, A. Bouhelier, L. Markey, J.-C. Weeber, C. Finot, and A. Dereux, Nano Lett. 9, 2935 (2009).
- [22] P. M. Bolger, W. Dickson, A. V. Krasavin, L. Liebscher, S. G. Hickey, D. V. Skryabin, and A. V. Zayats, Opt. Lett. 35, 1197 (2010).
- [23] I. P. Radko, M. G. Nielsen, O. Albrektsen, and S. I. Bozhevolnyi, Opt. Expr. 18, 18633 (2010).