https://doi.org/10.1051/epjconf/202328705035

Figure of merit for design of ε-near-zero metamaterials with enhanced Kerr-type nonlinearities

Francesca Dodici[1]*, *Domenico* Genchi[1], *Tiziana* Cesca[1], and *Giovanni* Mattei[1]

[1]Department of Physics and Astronomy, NanoStructures Group (NSG), University of Padova, via Marzolo 8, 35131 Padova, Italy

Abstract. Metamaterials are artificial media designed to display properies going beyond those of ordinary materials. Particularly interesting are ε-near-zero (ENZ) media with real part of the permittivity going to zero in a certain spectral range. Examples of ENZ metamaterials are metal dielectric multilayers, which allow to tune the position of the ENZ wavelength depending on their composition and which have been found to have enhanced Kerr-type nonlinearities, i.e. nonlinear absorption and nonlinear refraction. In this work we define a figure of merit for the design of multilayer metamaterials with strong Kerr-type nonlinearities and compare our predictions with both simulations and experimental results.

1 Introduction

Metamaterials are composite, artificial media designed to present unusual properties not found in ordinary materials, such as negative refractive index, optical cloaking or hyperlensing [1]. This is made possible thanks to their subwavelength structure, specifically designed in both composition and geometry to obtain the desired response in the metamaterial. Particularly interesting are ε-near-zero (ENZ) metamaterials in which the real part of the dielectric permittivity crosses zero at a wavelength called λ_{ENZ}. ENZ behavior is also naturally occurring in materials such as transparent conductive oxides, but their applications are limited because their λ_{ENZ} is fixed and usually in the near infrared. Conversely, in ENZ metamaterials such as metal-dielectric multilayer, i.e. periodic arrangements of alternating metallic and dielectric layers, the position of λ_{ENZ} can be tuned, according to the *effective medium approximation* (EMA), with a proper choice of the constituent materials and metal filling fraction [1]. These materials have attracted a lot of interest in the last decade in nanophotonics, nonlinear optics, plasmonics and material science for their distinctive and exclusive linear and nonlinear optical characteristics [2]. Among these properties we have the enhancement of nonlinear effects such as the optical Kerr effect (OKE), a third order effect manifesting in a dependence of the absorption coefficient and refractive index of a material on the incident intensity [3]. This dependence is quantified by the OKE parameters β and n_2, which are respectively the nonlinear absorption coefficient and the nonlinear refractive index. Nonlinear absorption can be exploited in the making of perfect absorbers, whereas nonlinear refraction can be useful for all-optical switching. In this work we focus on the optimization of the design of metal-dielectric multilayers to improve their Kerr-type nonlinear optical response.

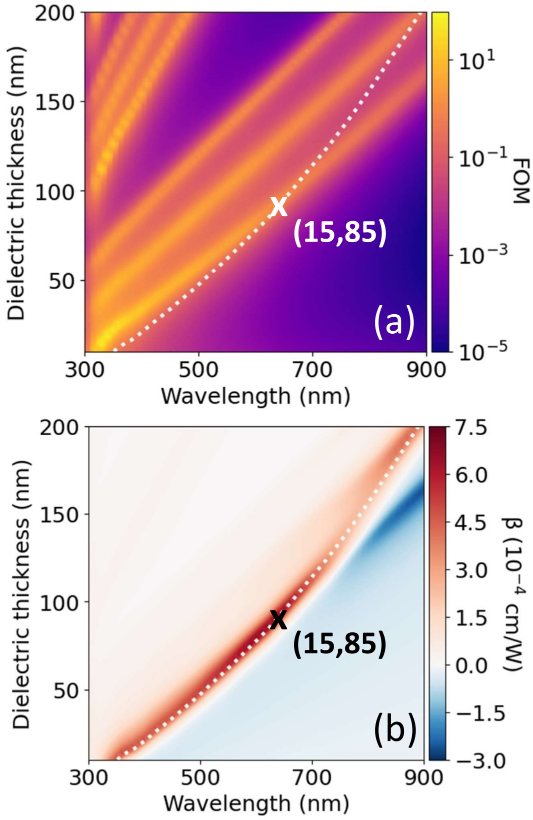

Figure 1. Comparison between the figure of merit (a) and nonlinear absorption coefficient β (b) calculated with the method described in [4] mapped as a function of the wavelength and dielectric layer thickness for a four period Ag/Al_2O_3 multilayer with silver layer thickness $t_m = 15nm$ and $t_d \in [10, 200]$ nm. The dotted lines highlight the value of λ_{ENZ}.

*e-mail: francesca.dodici@phd.unipd.it

2 Figure of merit

One of the most appealing features of metamaterials is the possibility to tailor their properties for specific applications with proper design of their structure and composition. In spite of their simple structure, several factors must be taken into consideration also with metal-dielectric multilayers. To begin with, in the interaction with an electromagnetic field, the generation of coupled plasmon polaritons at the metal-dielectric interfaces leads to transmission of light through thicknesses much larger than the metal skin depth and local intensity enhancement of the electric field in the metallic layers [5]. This local field enhancement is also responsible for the strong nonlinear response and can be quantified by considering an effective filling fraction ϕ_m as defined in [4]. Stronger nonlinearities are expected at larger values of ϕ_m, i.e. when the electric field localization in the metallic layers is highly increased. However, the presence of a metallic component, also implies losses caused by strong reflection at larger wavelengths and by absorption near the interband region of the metal. If these losses are significant, the output signal is reduced and so is the efficiency of the metamaterial in practical applications such as all-optical switching. Consequently, for an optimized design, we can define the following figure of merit (FOM) to find the combination of materials and thicknesses for which, at a certain λ, we have both strong field localization in the metal layers and low losses at the same time:

$$FOM = \frac{T}{R}\frac{\phi_m}{Im[\varepsilon_m]} \tag{1}$$

where T and R are respectively the transmittnace and reflectance of the multilayer, and $Im[\varepsilon_m]$ is the imaginary part of the metal permittivity. As an example, in Fig.1(a) we show the this FOM spectra for a 4 period Ag/Al_2O_3 multilayer with thickness of the silver layers fixed to t_m =15 nm and dielectric layer thickness t_d ranging from 10 nm to 200 nm. An equivalent map for the nonlinear absorption coefficient β of this multilayer metamaterial can be obtained using the model described in [4] and is shown in Fig.1(b). It can be seen that the FOM map presents distinct bands of maximum which correspond to the surface-plasmon modes supported by the metamaterial. On the other hand, for β, the strongest nonlinear response is found where the right-most band of the FOM is located. This is due to the proximity of this band to the ENZ regime (highlighted by the dotted lines in Fig.1), another condition known to boost the nonlinear response of the multilayer [6]. To see the effect of the constituent materials, the same analysis is carried out for Au/Al_2O_3 multilayers with similar results but lower value of the FOM.

3 Experimental methods and results

To check the validity of our predictions, we chose to fabricate by magnetron sputtering depositions, a four period Ag/Al_2O_3 multilayer in which λ_{ENZ} matches the FOM maximum. As shown in Fig. 1, this happens for (t_m, t_d)= (15/85) nm. The spectral trends of the OKE parameters $\beta(\lambda)$ and $n_2(\lambda)$ are measured with the z-scan technique and a peak in n_2 and β is found at λ_{ENZ}. The same results are obtained for the equivalent sample but with Au as metal instead of Ag. In this case however, as predicted by the lower value of the FOM, the nonlinear response at λ_{ENZ} is much smaller.

4 Conclusions

We defined a figure of merit to optimize the design of metal-dielectric multilayer metamaterials. This allows to evaluate the geometry and materials combination leading to the best overlap between having high local electric field enhancement and low optical losses in the ε-near-zero regime. Comparison with both simulations and experimental data showed good agreement with our predictions. Thus with this FOM, we introduced a useful predictive tool in the design of multilayers with optimized nonlinear optical response in the visible regime for possible applications in optical limiting or all-optical switching devices.

References

[1] P. Shekhar, J. Atkinson, Z. Jacob, Nano convergence **1**, 1 (2014)

[2] J. Wu, Z.T. Xie, Y. Sha, H. Fu, Q. Li, Photonics Research **9**, 1616 (2021)

[3] O. Reshef, I. De Leon, M.Z. Alam, R.W. Boyd, Nature Reviews Materials **4**, 535 (2019)

[4] D. Genchi, I.G. Balasa, T. Cesca, G. Mattei, Phys. Rev. Applied **16**, 064020 (2021)

[5] N.N. Lepeshkin, A. Schweinsberg, G. Piredda, R.S. Bennink, R.W. Boyd, Physical review letters **93**, 123902 (2004)

[6] S. Suresh, O. Reshef, M.Z. Alam, J. Upham, M. Karimi, R.W. Boyd, ACS Photonics **8**, 125 (2020)