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# Nonlocal effects in double fishnet metasurfaces nanostructured at deep subwavelength level as a path toward simultaneous sensing of multiple chemical analytes

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**Abstract**

Nanoplasmonic devices are among the most sensitive chemical sensors, with sensitivities reaching the single-molecule level. An especially convenient class of such sensors is that based on metasurfaces with subwavelength nanoholes, examples being extraordinary optical transmission arrays and double fishnet structures. Such structures ensure operation both in transmission and reflection mode and ensure high sensitivities and excellent coupling with external readout. In this paper we consider the possibility to tailor the response of aperture-based sensor structures by modifying the geometry of nanoholes at the deep subwavelength level through ensuring controlled use of nonlocal effects. We investigate the case where nonlocality is achieved by modifying the basic metamaterial fishnet structure (a metal-dielectric-metal sandwich with rectangular openings) by superposing additional subwavelength patterns, ensuring the appearance of new optical modes. The obtained unit cell superstructure will have multiple tailorable spectral peaks that will increase the selectivity at different wavelengths. The finite elements method was used for simulations of the proposed structures. As an example, we applied our results to the case of a benzene sensor, showing that its spectral properties and selectivity can be tuned by modifying geometry at a deep subwavelength scale. The obtained custom-designed spectral selectivity is convenient for multianalyte chemical sensing using a single structure.


*Keywords:* Nanoplasmonics; Double fishnet metamaterials; Metasurfaces; Chemical sensing; Subwavelength apertures; Electromagnetic nonlocality; Surface plasmons polaritons

## 1. Introduction

Plasmonic sensors [1,2] are among the most sensitive chemical or biological sensors, their sensitivities reaching the single molecule level [3]. They utilize the existence of surface plasmons polariton (SPP) modes along a conductor–dielectric interface to localize electromagnetic fields into volumes well below the diffraction limit. These sensors are basically refractometric. During measurement, they may be fully immersed into analyte utilizing some microfluidic scheme or only a thin layer of analyte may exist on their surface. In both cases the refractive index of the analyte will modify the propagating conditions of the SPP and thus generate the sensor response. Different geometries of plasmonic sensors are

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used, from simple metal slabs to complex nanoantennas [4,5].

One of the issues of interest in plasmonic sensing is the selectivity enhancement. Various methods are used to that purpose. Typically a layer of receptor substance is deposited onto the active surface of the sensor, ensuring adsorption of a target analyte [6]. Another interesting point is the possibility of simultaneous multianalyte detection. This can be obviously done by using an array of devices in parallel and ensuring multiplexing of their outputs [7]. However, a simpler approach is to use a single device that is inherently multiplexed. A method to ensure spectral multiplexing is to use a sensor with multiple resonance peaks at different frequencies, thus basically offering built-in spectral analysis.

Two large groups of nanoplasmonic sensors belong to the devices with ordered nanoaperture arrays. One of them is the group of extraordinary optical transmission structures [8,9], where a nanohole array is fabricated in a single opaque metal sheet and the presence of chemical/biological analyte modifies its transmission. Another group includes devices based on double fishnet metamaterials with negative refractive index [10,11]. In this group holes are fabricated in a metal-dielectric-metal sandwich, and the device again can operate in transmission mode. Both groups belong to aperture-based 2D plasmonic crystals [12].

A method to ensure multiple resonances at different wavelengths is to modify the basic geometry of the sensor. This can be done by fabricating superstructures obtained as superposition of simple ("primitive") forms. In the case of nanohole array-based sensor, this effect can be obtained by superposing two or more nanoholes with different geometries or dimensions within the same unit cell of the plasmonic crystal. Literature describes the use of small changes of unit cell shapes for tweaking of the spectral response [13,14]. A possibility to utilize Boolean combinations of primitive shapes to obtain complex subwavelength shapes has been considered in [15]. Albeit the changes are deeply subwavelength, they cause the appearance of new electromagnetic modes as a consequence of nonlocal response [16,17].

Nonlocality of the electromagnetic response is caused by the spatial dispersion of optical parameters at the subwavelength level. To avoid possible confusion, we stress that the nonlocality studied in this paper is observed in the context of the enhanced deviation of the optical response from the effective medium approximation [18], and does not consider electron–electron interactions and quantum description in subnanometer domains, like e.g. in [19].

Fig. 1. Fishnet structure (metal-dielectric-metal) without a superstructure array (left) and with a double superstructure with small (middle pattern) and large (right) shift of the basic pattern along both axes.

In this contribution we consider the possibility to engineer the optical nonlocality in our plasmonic structures and thus obtain a complex response with multiple resonance peaks at different wavelengths. To this purpose we utilize a simple approach where a primitive shape is superposed over itself with a certain shift, effectively corresponding to the Boolean AND operation in geometrical domain. The intention is to increase sensitivity in different parts of the spectrum and at the same time to ensure the use of a simple photolithography procedure. To design our structures we perform ab initio simulation of double fishnet metasurface utilizing Comsol Multiphysics finite element software package with and without deep subwavelength modification. We check the applicability of our approach on the example of ethanol–benzene mixture.

## 2. Method

A typical structure of fishnet metamaterial with negative effective refractive index in the optical wavelength range is shown in Fig. 1 left. It represents an array of square nanoholes in a metal-dielectric-metal sandwich (the middle dielectric is represented by lighter color).

Our idea is to superpose another array over the basic one, but with a shift along both the $x$ and the $y$ axis. As an example, Fig. 1 middle shows a structure where the identical array of square nanoholes is superposed to the basic pattern by simply shifting the original pattern along both in-plane axes by one quarter of the square hole side length, while Fig. 1 right shows the pattern obtained by a three-quarters shift.

The increased number of sharp edges in the superstructure on the right side of Fig. 1 will result in the appearance of a larger number of spectral peaks at different wavelengths, each connected with a different mode caused by the geometry change. Thus the obtained structure will allow for multispectral interrogation of its output signal and will offer a possibility to perform