Plasmonic nanostructures with local temporal response: a platform for time-varying photonics

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***Abstract*—This work is devoted to the development of an approach for implementation and designing time-varying media. A mechanism based on the use of plasmonic nanostructures with a reduced plasmon lifetime is proposed. It is shown that such nanostructures can be used to enhance the strength and speed of modulation of the refractive index of nonlinear media. This is achieved through decreasing of the spectral dispersion of the real permittivity. Plasmonic materials with peculiar optical properties, such as flat dispersion in the near-infrared range, were synthesized. For this purpose, we prepared TiON thin films and performed thermal post-treatment for fine-tuning permittivity of TiON. It has been shown that the proposed materials allow one to achieve an ultrashort plasmon lifetime on the order of 0.1 fs, which is an order of magnitude shorter than in the case of traditional plasmonic materials.**

***Keywords*—metamaterial, time-varying media, temporal inhomogeneity, plasmonics, tunable materials**

1. INTRODUCTION

Metamaterials open up wide opportunities for controlling optical fields, which makes them a promising platform for the development of innovative photonic devices. The unique optical properties of metamaterials are directly related to their spatial inhomogeneity. In recent years, a new class of artificial materials, called time-varying media, has attracted a broad interest [1]. In this case, the medium is homogeneous in space, but has temporal inhomogeneity. Materials with time-varying parameters are of great theoretical and practical interest. This is due to recent discoveries of a number of fascinating effects: violation of reciprocity, temporal aiming, amplification and compression of waves, etc. [1]. Temporal inhomogeneity is created by changing the optical parameters of the medium using an external influence. To observe the aforementioned effects, ultrafast modulation of the medium is required: the switching time must be less than the period of wave oscillations. For example, in the telecommunications range (near the wavelength of 1.5 μm), the wave period is approximately 5 fs. Therefore, there is a need for the practical implementation of materials with a sub-fs temporal inhomogeneity.

Temporal inhomogeneity is typically created using a nonlinear material whose refractive index is changed by pulsed laser excitation. In general, the switching speed is determined by the duration of the incident pulse and the parameters of the medium, namely, the time of the nonlinear response. The pulse duration of modern lasers can be much less than 1 fs, so the main limiting factor is the response time of the medium. A promising platform for implementation time-varying materials is epsilon-near-zero (ENZ) media [2]. These materials have a large and ultrafast sub-ps nonlinearity. However, ENZ media suffer from impedance mismatching. As a result, the incident field do not penetrate into the ENZ medium and cannot efficiently interact with it. This problem can be overcome by placing plasmonic nanostructures on the surface of the ENZ medium [2]. In this geometry, the optical pumping filed is the near-field of plasmonic nanostructures. Specifically, plasmonic structures re-emit the incident pulsed radiation. For this reason, the switching speed will be determined not by the duration of the laser pulse, but by the duration of the re-emitted one. In turn, the duration of the re-emitted pulse depends on the quality factor of the plasmon resonance. In recent years, great efforts in plasmonics have been directed to increasing the quality factor of the resonance. However, for the implementation of time-varying media, structures with a low Q factor are required. The lower the Q factor, the shorter the plasmon lifetime, and, therefore, the duration of the re-emitted pulse. Ultimately, this will lead to the increase of the switching speed in hybrid plasmonic-ENZ structures. This work is devoted to the development of plasmonic nanostructures with ultrashort plasmon lifetime.

2. RESULTS AND DISCUSSION

The lifetime of the localized plasmon in a subwavelength nanostructure is defined as follows [3]:

$$\tau = \left(\partial\left(\operatorname{Re}\varepsilon\right)/\partial\omega\right)\left(2\operatorname{Im}\varepsilon\right)^{-1}, \tag{1}$$

where Re(ε) and Im(ε) stand for real and imaginary parts of the underlying material, respectively. The magnitude of Im(ε) describes the level of optical losses. Thus, the decrease of the plasmon lifetime can be achieved by using materials with high absorption. However, this will lead to a decrease of the field enhancement factor.

Another way to reduce the plasmon lifetime is to use materials with low spectral dispersion of Re(ε) (see (1)). However, existing plasmonic materials are typically highly dispersive. There is a need for plasmonic materials with reduced spectral dispersion in the visible and near-IR ranges. To achieve this goal, we proposed the use of tunable plasmonic materials based on titanium oxynitride (general formula TiO_xN_y) [4]. A series of thin films (50 nm) of TiO_xN_y was synthesized using various conditions of magnetron sputtering. Then, a post-treatment procedure (thermal induced oxidization) was applied to the prepared TiO_xN_y films to fine-tune their dielectric function. This allowed us to achieve a peculiar optical property, such as flat spectral dispersion within the near-IR range (Fig. 1 (a)). For comparison, the spectral dispersion of real permittivity for the case of gold is on the order 0.1 nm^{-1} in the near-IR range, while for the case of prepared TiO_xN_y – on the order of 10^{-3} nm^{-1}. The optical properties of the synthesized materials were determined using spectral ellipsometry.

The derivative Re(ε) with respect to the wavelength (or frequency) determines the ability of a resonator to store electromagnetic energy. Consequently, as the dispersion approaches zero, the quality factor of the plasmon resonance vanishes. This can lead to the lack of plasmon-induced field enhancement. To analyze this effect, scattering cross section spectra of a subwavelength sphere were calculated using 3D electromagnetic solver (Ansys Lumerical). A finite-difference time-domain method (FDTD) was used. The diameter of the sphere was 10 nm. A total-field scattered-field source (400-700 nm), which illuminates the nanosphere with a plane wave, was used. Spectral dependence of permittivity values was input into the FDTD model and fitted using Lumerical's multi-coefficient model. The simulation volume was 2x2x2 μm^3. The non-uniform meshing algorithm was used. The cells size in the region surrounding the sphere was 0.5 nm, and for regions far from the sphere the cells size was set to 10 nm. The time step was 6x10^{-4} fs. Perfect matching layers boundary conditions were used in all directions. Fig. 1(c) shows the scattering cross section spectra calculated for two materials with different dispersion values (Fig. 1(b)). For proper comparison, the optical losses of both materials are set the same ($Im(\varepsilon) = const \approx 0.5$) within the considered spectral range. The dielectric functions are chosen so that the plasmon resonance is near the wavelength of 550 nm. As can be seen from Fig. 1(c), the maximum value of the scattering cross section for both materials is the same. This suggests that the enhancement factor at the resonant wavelength is also the same. Thus, a decrease of dispersion does not lead to a decrease in the field enhancement factor. This can be understood as follows. When the dispersion is decreased, less energy is stored by the resonator, but the oscillation amplitude increases. The latter is due to the fact that in the absence of dispersion, the electronic response becomes local in time: the inertia of electrons decreases.

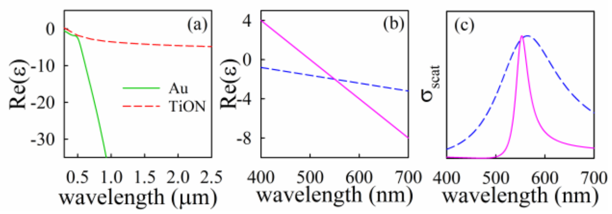

Fig. 1. (a) Plots of the real part of the dielectric function for gold (solid line) and synthesized TiON film (dashed line). (b) Plots of the real part of the dielectric function and (c) corresponding scattering cross section spectra of a spherical nanoparticle. The data in (b) and (c) are given for two types of materials: with high (solid line) and low (dashed line) dispersion

3. CONCLUSIONS

In this work, we proposed a mechanism for reducing the plasmon lifetime by an order of magnitude compared to traditional plasmonic materials. At the same time, the ability of the plasmonic structure to enhance the field is retained. This effect is achieved by reducing the dispersion of the real part of the permittivity. A TiON-based plasmonic material with flat dispersion in the near-IR range was synthesized. The results of this study open up the possibility of creating time-varying media with a large depth and speed of modulation. This will increase the efficiency of interaction of optical fields with temporal interfaces.

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