Efficient Optical-Harmonics Generation and Nonlinear Purcell Effect in Metal/Photonic Crystal Structures

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Abstract: Enhancement of second and third optical-harmonics generation is experimentally observed in metal/photonic crystal structures under conditions of Tamm plasmon-polariton excitation. It is shown that amplification can occur via two mechanisms: resonant fundamental wave and resonant second-harmonic.

1. Introduction

Tamm plasmon-polaritons (TPP) are optical surface states which appear as electromagnetic field localization at an interface of photonic crystal (PC) and metal film [1]. TPPs do not have phase-matching conditions for in-plane wave vector, and can be excited for any angle of incidence and polarization of incoming radiation [2]. Experimentally these states manifest themselves as narrow resonances in transmittance or reflectance spectra of Me/PC systems. TPPs can be effectively coupled with other localized states, such as microcavity modes [3], excitons [4], and surface plasmons [5], which have been used to create compact lasers and sensors [6,7].

Imaging and sensing with optical-harmonics (OH) is widely used for biological objects and buried interfaces, since nonlinear-optical phenomena are extremely sensitive to boundaries and interfaces. Localization of fundamental field inside nonlinear medium leads to an enhancement in OH intensity. In PCs such phenomenon appears at the edge of a photonic bandgap [8], in metals OHG is enhanced in the presence of propagating or localized surface plasmons [9].

It was shown that for specially designed PCs, one can overlap edge of a fundamental bandgap with a fundamental wave, and edge of a third-order bandgap with a third-harmonic wave, which results in an additional amplification of the optical-harmonic generation efficiency [10].

2. Samples and setup

Samples of metal/photonic crystal structures were prepared using thermal evaporation and magnetron sputtering techniques. Two sets of samples were manufactured for experiments on second- and third-harmonic generation, respectively. First-type samples consist of 7 SiO_2/Ta_2O_5 bilayers, with thicknesses of 92 nm for Ta_2O_5 and 130 nm for SiO_2. They are covered with a semitransparent 30-nm-thick silver film, passivated with a 10-nm-thick aluminum oxide film. TPP resonant wavelength in first-type samples is at 820 nm under normal incidence. Second-type samples were designed using transfer matrix technique to fulfill double-resonance conditions for Tamm plasmon-polaritons in fundamental and third-order photonic bandgaps. They are composed of 5 $SiO_2/Ta_2O_5/SiO_2/Ta_2O_5$ quad-layers with thicknesses of 240/164/284/194 nm, respectively, covered with a 28 nm of silver and a passivation Al_2O_3 layer. Fundamental TPP resonance appears at 1580 nm under normal incidence.

Femtosecond laser (Ti:sapphire for first- and Er:fiber for second-type samples) was used as a source of fundamental radiation, which passed through two Glan-laser prisms, allowing for control of power and polarization, and was focused onto the sample. Sample was mounted on the rotational stage, providing 0.001° accuracy of an angle of incidence.

3. Experimental results

Enhancement of the second-harmonic generation in case of the fundamental field localization in metal/PC structure was previously reported in [11]. In this study we focus on the case when the fundamental field is not in resonance with any of the eigenstates of a metal/PC structure, whereas the frequency of the second-harmonic radiation is in resonance with TPP. The nonlinear analogue of the Purcell effect leads to the enhancement of the stimulated emission to the second-harmonic, thus increasing effective nonlinear susceptibility of the metal/PC sample. We have shown that normalized conversion efficiency in case of the resonant second-harmonic radiation can be 25-fold enhanced in the studied sample in comparison with a bare silver film, and reaches 5×10^{-11} W^{-1}.

The main result of the study is the realization of double-resonance conditions for Tamm plasmon-polaritons, when the wavelength of the fundamental TPP mode is exactly the tripled frequency of the third-order TPP. Under such conditions, optical-harmonics generation process can benefit from both the fundamental field localization inside the nonlinear structure and the nonlinear Purcell effect, which enhances emission at the third-harmonic wavelength. Special design of the second-type structure, described in Sec.2 was developed to overcome dispersion effects, which usually detune higher-order TPP frequencies from the multiples of the fundamental one. One of the possible parameters, which effectively controls the overlapping of fundamental and third-order resonances and, at the same time, don't change the amount of nonlinear material too much, is the thickness of the topmost layer of a PC, adjacent to the metal. Figure 1 shows results of the numerical calculation of the normalized third-harmonic generation efficiency versus the angle of incidence and the thickness of a topmost layer of a PC. Grey curves correspond to dispersion curves of fundamental and third-order TPPs. TH conversion efficiency is enhanced in the vicinity of the fundamental TPP resonance due to field localization inside metal/PC structure and reaches 0.5×10^{-9} W^{-2}. Enhancement due to nonlinear Purcell effect in the third-order TPP mode is rather weak, however when dispersion curves overlap (at 19° angle of incidence and thickness of the topmost layer of 194 nm), this process adds up to the aforementioned one and the resultant conversion efficiency peaks at 2×10^{-8} W^{-2}. Sample, used in an experiment has the same parameters as in the calculations, which was proven by SEM and ellipsometry. Experimental data have shown that THG is enhanced by the factor of 6×10^4 in metal/PC sample in comparison with a bare silver film, which is in an agreement with the results of numerical calculations.

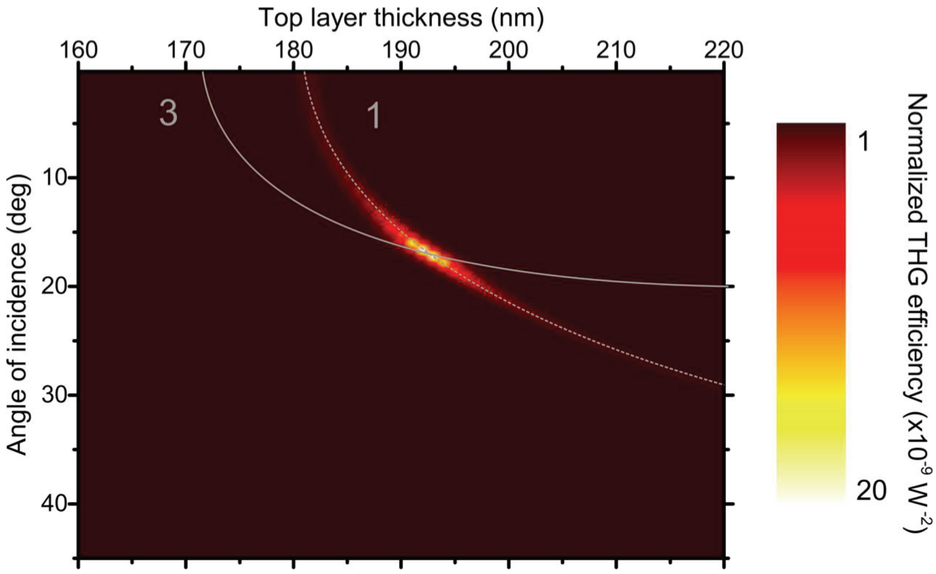

Fig.1. Efficiency of THG in PC/Me sample for *pp* combination of fundamental and TH radiation polarizations, versus the angle of incidence and thickness of the topmost layer of a PC. Gray curves are guides for the eye and represent dispersion curves of the fundamental (1, dashed) and third-order (3, solid) Tamm plasmons.

4. Conclusions

We have shown that in metal/photonic crystal structures optical-harmonics generation is enhanced under conditions of Tamm plasmon-polariton excitation. Two mechanisms are studied: fundamental wave localization in nonlinear layers and resonant tunneling of optical-harmonic radiation through Tamm plasmon mode. Enhancement of conversion efficiency varies from one to two orders of magnitude in single-resonance cases, while conversion efficiency from first to third harmonic is enhanced by four orders of magnitude and reaches 2×10^{-8} W^{-2} when both mechanisms are present in a sample with a special design.

5. References

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