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Towards harmonic generation enhancement on silicon

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Abstract. Nowadays, nanostructures are routinely fabricated and integrated in different photonic devices for a variety of purposes and applications. For instance, nonlinear silicon photonics is an area of interest due to its high compatibility with CMOS technology, offering structure sizes down to 10nm at low cost. When the nanoscale is reached, light-matter interactions can display new phenomena, conventional approximations may not always be applicable, and new strategies must be sought in order to study and understand light-matter interactions at the nanoscale. In this work, we report a comparative experimental and theoretical study of second and third harmonic generation from silicon with the aim of explaining the nonlinear optical properties of this material at the nanoscale. We measure second and third harmonic efficiencies as a function of angle of incidence, polarization and pump wavelength. We compare these measurements with numerical simulations based on a microscopic hydrodynamic model which accounts for different possible contributions to the nonlinear polarization. This way, we have the ability to explain properly the SH and TH signals arising from different silicon samples. Once we have this knowledge, we are able to design more complex structures, such as silicon nanowires, where higher conversion efficiencies can be achieved.

1 Introduction

Second and third harmonic generation (SHG and THG) are two nonlinear processes in which when light interacts with a nonlinear material, new frequencies are generated. These processes are typically studied in thick materials with high nonlinearities and under phase matching conditions and low material absorption with the aim of achieving high conversion efficiencies. They are described through the nonlinear polarization, which is usually approximated to the electric dipole contribution described by the second and third order susceptibilities $\chi^{(2)}$ and $\chi^{(3)}$. This approximation starts to fail when the nanoscale is reached: SHG and THG conversion efficiencies are drastically reduced and phase matching conditions and absorption no longer play a significant role. In this case, other nonlinear contributions to the nonlinear polarization, such as magnetic dipole and electric quadrupole interactions, must be taken into account.

In this work, we analyse SH and TH fields generated when ps and fs pulses tuned at 1064nm or 800nm, respectively, interact with different silicon samples. This analysis consists on first, measuring the harmonic signals as a function of incident angle, polarization and pump wavelength, and then, comparing them with numerical predictions based on a microscopic hydrodynamic model explained in details in [1]. The study starts by analysing simple structures: an amorphous silicon nanolayer and a crystalline silicon wafer. The comparison of measurements with numerical simulations allows us to obtain information of the material and the light-matter interaction at nanoscale. With this knowledge, we are able proceed the study by designing more complex structures, such as silicon nanowires, where higher conversion efficiencies can be achieved.

2 Results

2.1. Experiments

Measurements of SH signals from silicon have been conducted in transmission and in reflection. In both cases, incident pulses tuned at 800nm and 1064nm have been used. The process of THG was conducted either in transmission and reflection using incident pulses tuned at 1064nm. For this purpose, we have developed an experimental set-up capable of analysing the angular dependence of the harmonic signals in both transmission and reflection configurations, and as a function of polarization and pump wavelength. This set-up also allows us to properly separate the harmonic fields from each other and from the fundamental field radiation by means of filters and a dispersive prism.

2.2 Theory

We have performed numerical simulations to model the experimental situation. The theoretical model embraces full-scale time-domain coupling of matter to the

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macroscopic Maxwell's equations. We use the hydrodynamic model to understand linear and nonlinear optical properties of different materials by accounting for competing surface, magnetic and bulk nonlinear sources, which in this case consist of mostly bound electrons. In semiconductors like silicon, the action of free carriers is neglected, so that the dielectric function of the material can be described through the Lorentz model containing one resonance in the UV range. We measure and estimate the efficiency of the harmonic signal using no assumptions about effective surface or volume nonlinearities other that effective electron masses and densities.

In Fig. 1 we show results of reflected SHG efficiencies as a function of incident angle from a 25nm-thick amorphous silicon nanolayer. Pump pulses tuned at 1064nm were used and both incident and harmonic fields were TM-polarized. Measurements are plotted with full circles while the solid curve corresponds to the predicted efficiencies. As it can be seen, experimental and simulated curves agree remarkably well, showing a maximum predicted and detected efficiency at 50º incident angle.

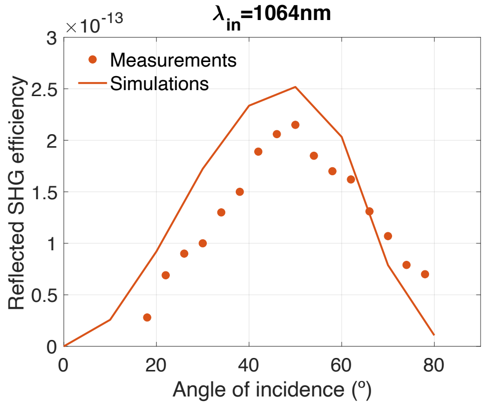

Fig. 1. Measured (full circles) and predicted (solid curve) reflected SHG efficiencies as a function of incident angle from a 25nm-thick amorphous silicon layer when the pump pulse is tuned at 1064nm and both fundamental and SH fields are TM-polarized.

In Fig. 2 we show experimental and predicted results of transmitted and reflected TM-polarized SHG efficiencies as a function of incident angle from a 25nm-thick amorphous silicon layer when an incident field tuned at 800nm and TM-polarized was used. We observe that the reflected experimental and theoretical curves agree well in shape and amplitude, having a maximum around 50º angle of incidence. The results for the transmission case are also satisfactory. Another thing that is reproduced in both experimental and theoretical results is the ratio between transmitted and reflected efficiencies. As a result, we are confident that our theoretical approach suffices to understand and explain most of the relevant aspects of surface SHG.

In Fig. 3 we show the results of the angular dependence of reflected THG efficiencies from a 500μm-thick crystalline silicon wafer. Pump pulses tuned at 1064nm were used. A TM and a TE-polarized TH field was detected for TM and TE-polarized incident light, respectively. Once again, both experimental and theoretical results agree remarkably well in shape and amplitude showing a maximum efficiency at normal incidence. The values of these efficiencies were used to retrieve the nonlinear dispersion of the material $\chi^{(3)}$, as reported in [2].

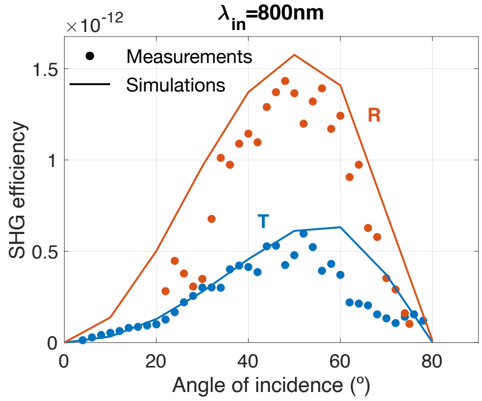

Fig. 2. Transmitted (blue) and reflected (red) measured (full circles) and predicted (solid curves) SHG efficiencies as a function of incident angle from a 25nm-thick amorphous silicon layer when the pump pulse is tuned at 800nm and both harmonic and fundamental fields are TM-polarized.

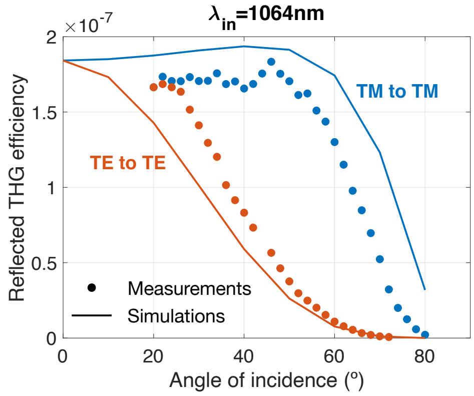

Fig. 3. Transmitted (blue) and reflected (red) measured (full circles) and predicted (solid curves) THG efficiency as a function of incident angle from a crystalline silicon wafer when the incident field is tuned at 1064nm. A TM (blue) and a TE-polarized (red) TH field was detected for TM and TE-polarized incident light, respectively.

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