Second-harmonic generation based on symmetry breaking in a periodically layered medium

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We demonstrate second-harmonic generation in a periodically layered medium made by atomic layer deposition. The layer sequence is chosen such that the inversion symmetry of the overall composite material is broken and a second-harmonic wave is allowed to build up.

Ever since the first demonstration of second-harmonic generation (SHG) in 1961 by Franken et al. [1], materials possessing a second-order susceptibility ($\chi^{(2)}$) are being investigated and used in a multitude of applications. They give us the possibility to create light in new wavelength regions through SHG, sum or difference frequency generation, and they can be used to fabricate optical parametric oscillators and electro-optic modulators (based on the Pockels effect). However, integrating these functions on a silicon photonics platform remains difficult. Indeed, silicon and (amorphous) silicon nitride are materials possessing inversion symmetry, which means $\chi^{(2)}$ vanishes in the electric dipole approximation. In silicon this problem has been circumvented by depositing a highly stressed silicon nitride layer on silicon waveguides [2,3]. This creates an inhomogeneous strain in the silicon crystal and breaks the inversion symmetry. Recently, researchers have demonstrated that also silicon nitride possesses a non-zero $\chi^{(2)}$ whose magnitude can be increased by tuning the material composition [4,5]. The origin of the second-order nonlinearity is still unclear. Naturally, each interface also breaks the inversion symmetry, which makes SHG a very useful tool to probe surfaces [6]. Here, we enhance this effect by creating a periodically layered medium consisting of many subsequent interfaces. Three materials A, B and C are periodically stacked (ABCABC…) to form an overall composite material without inversion symmetry. These materials are deposited by ALD (atomic layer deposition), which makes it easy to integrate them on silicon (nitride) nanophotonic structures [7].

Our samples consist of periodically deposited thin layers (0.7 nm) of TiO_2 (A), Al_2O_3 (B) and In_2O_3 (C) on top of a borosilicate glass substrate (SCHOTT BOROFLOAT® 33). Four samples were prepared: one with a total thickness of 2.1 nm, 25 nm, 50 nm and 75 nm. Plasma enhanced ALD was used for deposition: an oxygen plasma is created with a radiofrequency plasma source of 13.56 MHz and a power of 200 W. The ALD process is done by alternating pulses of metalorganic precursor at 6×10^{-3} mbar and oxygen plasma at 1.2×10^{-2} mbar. The substrate temperature is held constant at 120°C. The metalorganic precursors and growth per cycle for each of the three materials are respectively Tetrakis(dimethylamido)titanium(IV) and 0.06 nm/cycle for TiO_2, Trimethylaluminium and 0.1 nm/cycle for Al_2O_3, and Tris(2,2,6,6-tetramethyl-3,5-heptanedionato)indium(III) and 0.01 nm/cycle for In_2O_3.

In our experimental setup we use a pulsed Ti:Sapphire laser at a wavelength of 980 nm (Mai Tai® HP from Spectra-Physics). The pulse width is 100 fs and the repetition rate 80 MHz. We are working at an average power of 1.11 W. Parabolic mirrors (focal length of 50.8 mm) are used to slightly focus the beam onto the sample. After spectral filtering the second-harmonic power is measured with a femtowatt detector (measurement is done in transmission). The sample is placed on a rotation stage so the incidence angle can be varied. The polarization direction is controlled with a half wave plate. We use p-polarized light in our experiments.

The refractive index of our samples was determined from ellipsometric measurements. The ordinary refractive index is 2.02 (2.13) at 980 nm (490 nm) and the extraordinary refractive index is 1.92 (2.06) at 980 nm (490 nm).

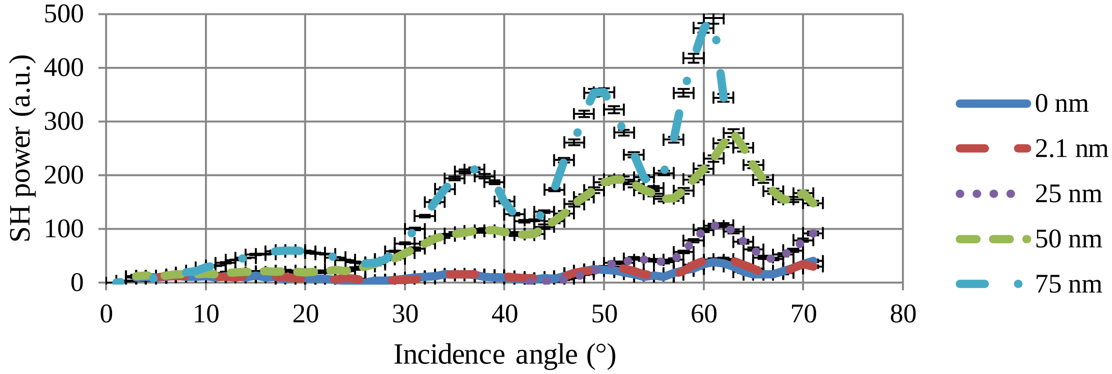

Fig. 1: Second-harmonic power measured in function of the incidence angle of the laser beam. 0° corresponds to normal incidence. The results are shown for a blank glass substrate (0 nm), for an ABC stack with a total thickness of 2.1 nm, 25 nm, 50 nm and 75 nm.

Figure 1 shows the results of our measurements. For normal incidence the detected SH power goes to 0, as the effective $\chi^{(2)}$ becomes 0. Even for the blank substrate we detect SHG, due to the surface $\chi^{(2)}$ of the glass. For increasing thicknesses of the ABC stack, and thus increasing number of interfaces, we measure increasing SH powers. This clearly indicates the presence of a bulk second-order nonlinearity in our periodically layered medium and shows that the SH radiation is not merely created at the air/ABC stack and ABC stack/glass interfaces. We estimated the major tensor component χ_{zzz} of the ABC composite material to be 0.4 pm/V. The fringes in the curves can be explained by the interference between the SH wave created in the ABC stack and the SH wave created at the back surface of the glass substrate. The visibility of the fringes is limited due to temporal walk off between the pulses.
In conclusion we can say that the breaking of the inversion symmetry in a periodically layered medium allows the generation of second-harmonic waves. We managed to reach a second-order susceptibility of 0.4 pm/V.

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