New Nanoplasmonic Devices and Fabrication for Large Enhancement and Tunability of Second Harmonic Light Generation

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Second harmonic generation (SHG) of light has broad and significant applications in communications, manufacturing, sensors, solar cells, displays, medical treatments, and many others. Two central issues in SHG are the enhancement and tuning [1-3]. Previously, SHG enhancements are low and tunability is still poor, which limit their real applications. Also, it was believed that the tuning of second harmonic generation by applying a voltage bias is due to third order electric susceptibility [4]. Here we propose and demonstrate a new nanoplasmonic device, termed "plasmonic charge assisted second harmonic generator (p-CASH) with high SHG enhancement of 76 fold (7,600 %), high tunability (7 % per volt), and wide tuning range (up to 280 %). Furthermore, we demonstrate that the SHG tuning of our p-CAHS device using voltage bias does not involve the effect of third order susceptibility, but rather strictly from the second order susceptibility.

The p-CASH is an asymmetric parallel-plate capacitor with thin layers of a plasmonic nanostructure, termed "disk-coupled dots-on-pillar antenna array" (D2PA)"[5], a 100 nm thick TiO2 (semiconductor and nonlinear) and a 500 nm SiO2 (insulator) sandwiched between two electrodes (the transparent indium-titanium-oxide (ITO) layer and the Au backplane of D2PA (Fig 1).

The p-CASH was fabricated by combining nanoimprint, self-alignment, and guided self-assembly, plus PECVD, E-beam evaporation and sputtering to deposit TiO2, SiO2 and ITO layers, and photolithography to define electrical contacts.

Figure 2(a) shows the measured second-harmonic generation (SHG) spectrum for pumping using 800 nm laser for three different systems. SHG intensity of p-CASH at 40V and 0V forward bias is 76 and 26 fold higher, respectively, than that of an identical TiO2 thin film on a glass. In addition, the conversion efficiency of our p-CASH is over 30 folds higher compared with previous tunable SHG plasmonic device [4].

Figure 2(b) shows the tuning of SHG of p-CASH using a voltage bias. At the forward bias, the SHG increases with the applied voltage (7 % per volt & 280 % tuning range) first and then saturates. At reverse bias, no change with the SHG signal was observed. Such bipolar behavior plus the Capacitance-Voltage measurements show that the SHG tuning is caused by the change of second order nonlinear susceptibility through charge injection, rather than the change of third order susceptibility through external static electric field, as believed before.

Moreover, the p-CASH is scalable to large area (wall-paper size) and easy to be fabricated massively (e.g. use of nanoimprint). Hence it not only provides new understanding of SHG tuning and high enhancements using novel nanoplasmonic devices, but also potentially opens up many new applications in various fields.

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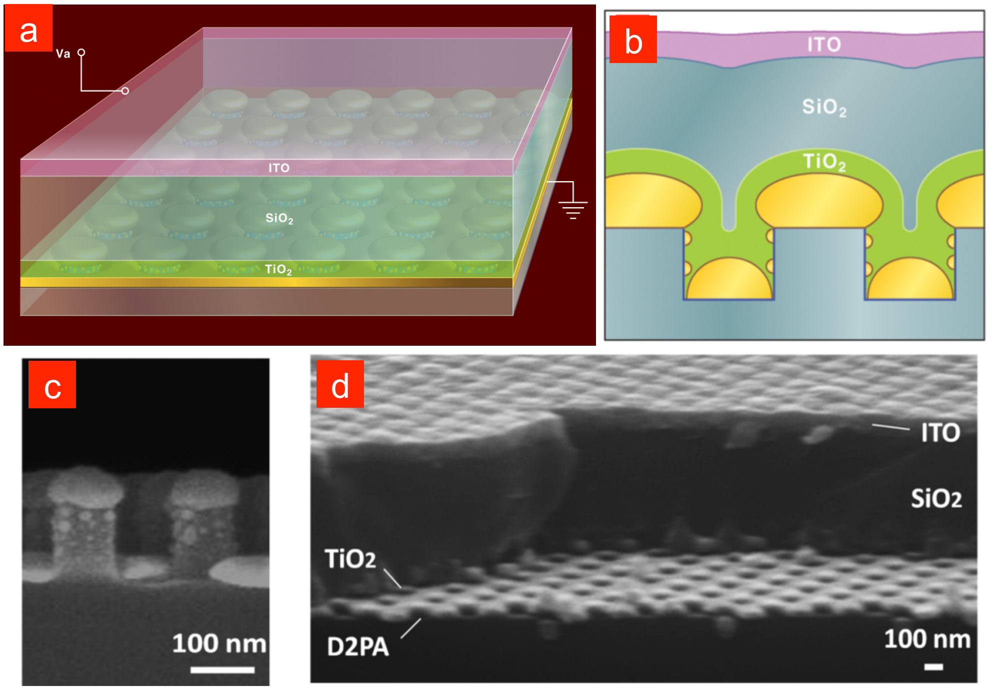

Fig. 1. p-CASH (plasmonic charge assisted second harmonic generator), a bipolar plasmonic structure. (a) Schematic. The p-CASH is an asymmetric parallel-plate capacitor with thin layers of a plasmonic nanostructure, a TiO2 (semiconductor and nonlinear) and SiO2 (insulator) sandwiched between two electrodes. (b) Cross-sectional schematic. And cross-sectional SEM of (c) the plasmonic substrate (D2PA) and (e) p-CASH.

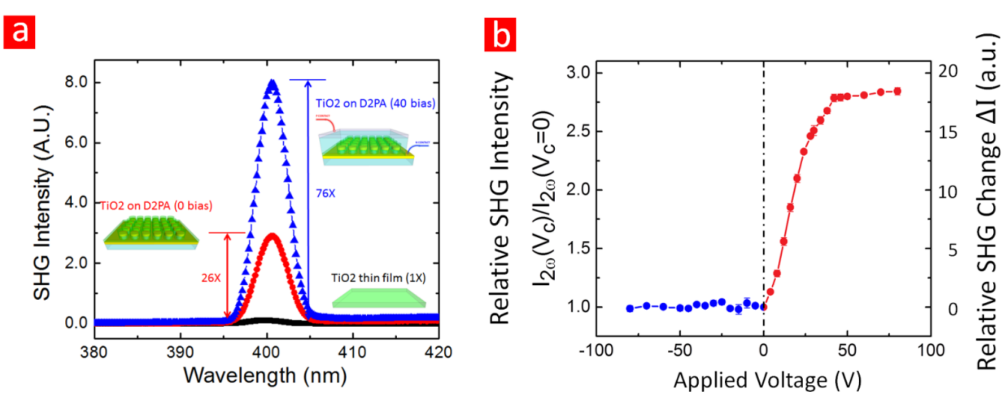

Fig. 2. (a) Second-Harmonic Generation spectrum for pumping 800 nm light for three systems: the p-CASH at 40V and 0V forward bias is 76 and 26 fold, respectively, higher than an identical TiO2 thin film on a glass. And (b) Double-Y plot showing the relative SHG intensity ($I_{2\omega}(V_c)/I_{2\omega}(V_c = 0)$) and the relative SHG change ($I_{2\omega}(V_c) - I_{2\omega}(V_c = 0)$) as a function of the voltage bias across the p-CASH.