# Enhanced second harmonic generation in individual barium titanate nanoparticles driven by Mie resonances

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### S1. Mode volume of all-dielectric nanoparticles

In all-dielectric nanoparticles, the electric resonances are derived from the oscillation of polarization charges, while the magnetic resonances are originated from the circular displacement currents that are excited inside the particle by incident light. The cavity mode effective volume can be calculated according to the formula (Kartik Srinivasan, *et al. Opt. Express* 2006, Cavity Q, mode volume, and lasing threshold in small diameter AlGaAs microdisks with embedded quantum dots):

$$V_{\text{eff},E} = \frac{\int_{V} \varepsilon(\mathbf{r}) |\mathbf{E}(\mathbf{r})|^{2} d^{3}\mathbf{r}}{\max\left[\varepsilon(\mathbf{r}) |\mathbf{E}(\mathbf{r})|^{2}\right]}$$

where  $\varepsilon(\mathbf{r})$  is the dielectric constant,  $|\mathbf{E}(\mathbf{r})|$  is the electric field strength, and *V* is a quantization volume encompassing the resonator and with a boundary in the radiation zone of the cavity mode under study. The electric mode is limited to interface while the magnetic mode exist inside the nanoparticle, so the latter has a bigger volume. We have added this content in the introduction. This question is clarified by Y. S. Kivshar's group (*Nano Lett.* 2014, Enhanced third-harmonic generation in silicon nanoparticles driven by magnetic response.) and J. Valentine's group (*Nano Lett.* 2015, Nonlinear Fano-resonant dielectric metasurfaces). The magnetic mode refers to magnetic dipoles and quadrupole. In order to understand better, we simulated the scattering of an individual silicon nanoparticle with a radius of 100nm.



**Figure S1**: The field distribution at resonances. ED: electric dipole; MD: magnetic dipole; EQ: electric quadrupole; MQ: magnetic quadrupole.

## S2. Excitation spectrum



**Figure S2.** Excitation spectrum emitted from Ti: sapphire femtosecond laser (Maitai HP) at 850 nm.

## S3. Backward scattering spectra



**Figure S3.** Backward scattering spectra of an individual BaTiO<sub>3</sub> sphere (410.0 nm) and a dimer (254.2 & 150.1 nm) experimentally and their corresponding simulations calculated by FDTD method. Scale bar, 200 nm.

S4. Refractive index of BaTiO<sub>3</sub>



Figure S4. Experimentally measured and theoretically calculated refractive index of  $BaTiO_3$ . *n* and *k* represent the real and imaginary parts of the index.



#### S5. Field distributions

**Figure S5.** Magnetic field distributions far from resonance region of  $BaTiO_3$  nanospheres calculated by FDTD method. (a) d=151.0 nm, (b) d=224.9nm