Supporting information for Second-harmonic generation from metal nanoparticles – resonance enhancement vs. particle geometry

Robert Czaplicki[†], Jouni Mäkitalo[†], Roope Siikanen[†], Hannu Husu[†,‡], Joonas Lehtolahti[§], Markku Kuittinen[§], and Martti Kauranen[†]

[†] Department of Physics, Tampere University of Technology, P. O. Box 692, FI-33101 Tampere, Finland

[‡] nLIGHT Corporation, Sorronrinne 9, FI-08500 Lohja, Finland

[§] Institute of Photonics, University of Eastern Finland, P. O. Box 111, FI-80101 Joensuu, Finland

Corresponding author: robert.czaplicki@tut.fi

Details of linear spectra

The linear spectra of the samples, presented in Figure 2 of the main text, were determined by measuring their extinction at normal incidence for *x* and *y* polarizations. In order to facilitate the interpretation of Figure 2, the details of the measured resonances are shown in the Table S1. Note that the important resonances (highlighted) have very similar linewidths, although they differ somewhat in their strengths. In addition, the detuning of the laser from the line center is relatively similar for each resonance.

Table S1. Details of the resonances. The highlighted data correspond to the resonances close to the laser wavelength used in the second-harmonic experiments.

Sample	*x*-polarized resonance				*y*-polarized resonance			
	Resonance wavelength [nm]	FWHM [nm]	Detuning from the 1060 nm	Optical density	Resonance wavelength [nm]	FWHM [nm]	Detuning from the 1060 nm	Optical density
L	1494	180	-	1.231	1023	92	37	1.019
T_b	1096	97	36	1.143	1611	244	-	1.515
T_s	1105	104	45	1.19	1023	100	37	0.68

Simulation of second-harmonic generation

In order to support the results obtained in the experiments, both the linear and nonlinear responses from the samples were simulated using the boundary element method (BEM).

Reliable simulation of the second-harmonic generation (SHG) signals requires very good agreement between the measured and calculated polarized extinction spectra. Although the overall agreement in our case is very good even on a quantitative level, small differences do appear at the fundamental wavelength used in the SHG experiments (1060 nm). These differences are particularly important for the sample T_s, where both *x* and *y* polarizations are near resonant (Figure S1).

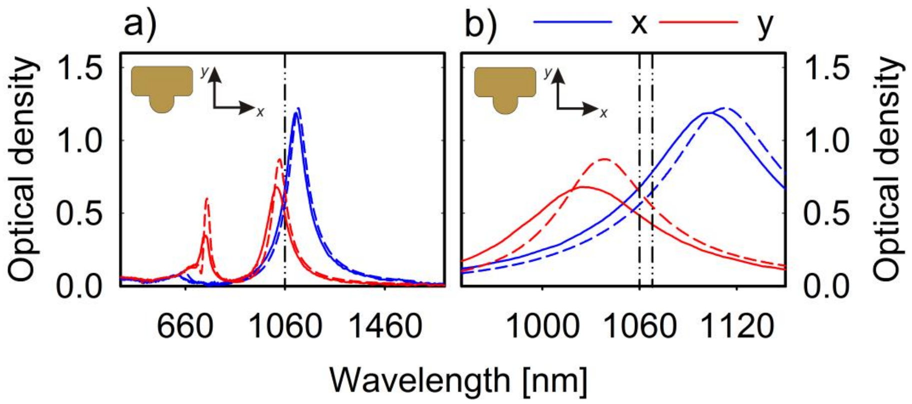

Figure S1. Polarized extinction spectra of sample T_s: a) full spectrum b) narrower range to show details of resonances and optical density levels at both used wavelengths. The solid lines denote measured spectra and dashed lines the calculated ones. The fundamental wavelength of our laser (1060 nm) is marked as vertical dash-dotted line. The second dash-dotted line in part a) shows wavelength (1068 nm) used for repeated calculations.

In order to show how such minor differences can influence the simulated SHG, we repeated the calculations for the wavelength (1068 nm) which represents qualitatively similar relative optical densities in simulations as measured in the experiment for our laser wavelength (1060 nm). The SHG signals simulated for 1068 nm fundamental wavelength (Figure S2) show even better agreement with the experimental data.

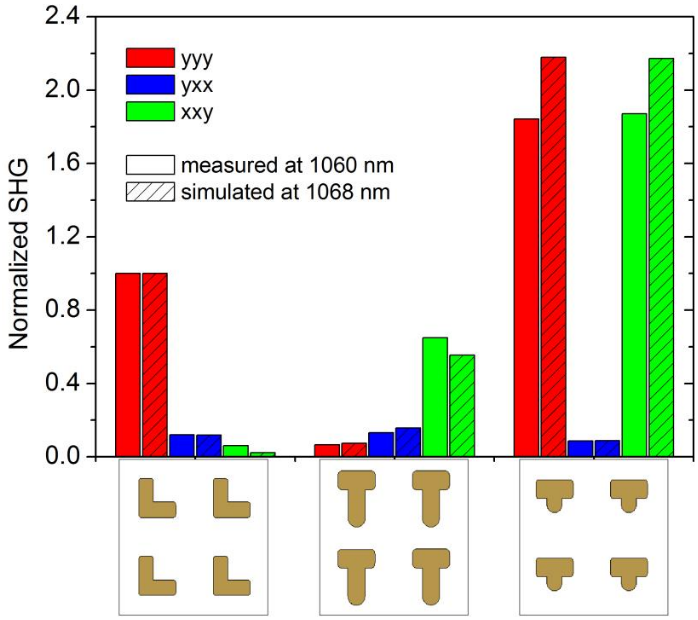

Figure S2. SHG signals from studied samples normalized to *yyy* component of sample L. The non-patterned bars represent experimental data (fundamental wavelength 1060 nm) and the patterned bars show calculated signals (fundamental wavelength 1068 nm).