Supporting Information

Third-Harmonic Generation Enhancement by Film-Coupled Plasmonic Stripe Resonators

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Normalization of experimental THG data points:

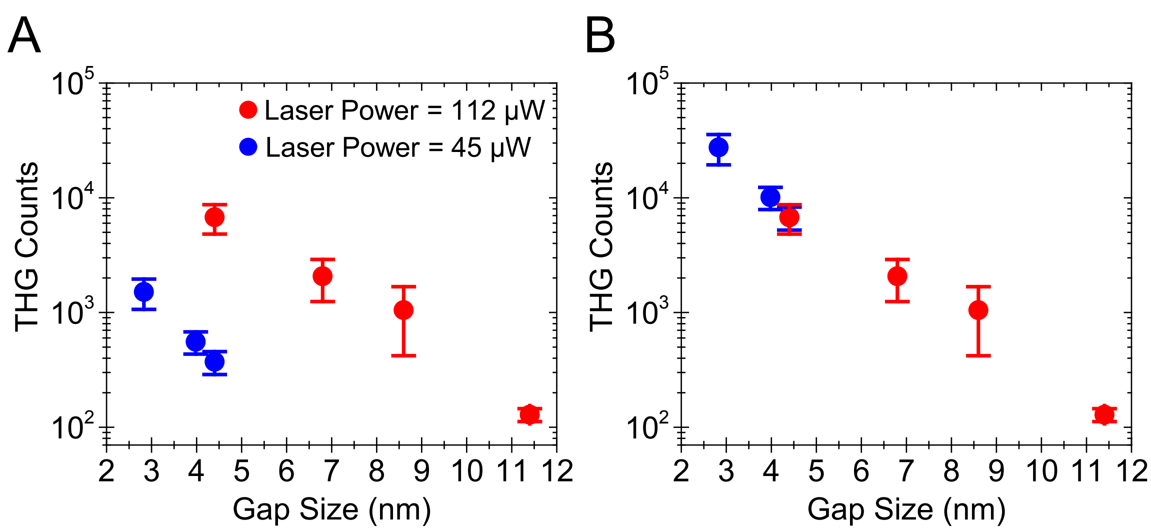

Figure S1: Third harmonic generation from stripe samples vs gap size. The data presented in this figure are the same as the experimental data reported in figure 4a, but here the signal intensity is reported in units of measured counts instead of enhancement. (A) Actual measured THG signal intensities for each sample. Red data points represent data taken using an input average laser power of 45 μW while blue data points represent data taken using an input average laser power of 112 μW. (B) Normalized THG signal intensities. Here the blue data points have been scaled by a factor of 18.19 such that the two data points at gap size = 4.40 nm are equal. The full justification for this normalization is presented in the text below.

The experimental third harmonic generation data for the samples with smaller gap sizes (g = 2.83 nm, g = 3.98 nm, and g = 4.40 nm) must be normalized, as depicted in Figure S1, in order to be correctly interpreted. The normalization is necessary because the two samples with the smallest gap sizes (blue data points) suffered from significant damage (determined by decreasing THG signal over time) when exposed to the laser at the same power level as was used for collecting data on the larger gap samples (red data points). We expended significant effort to find a laser power that could be exposed to all six samples without causing damage, but we were unable to find such an optimal laser power. As a result, we had to split the measurement into two different power regimes for the different samples in order to be able to carry out the experiment and normalize between them. In order to do this, we measured the THG signal of the g = 4.40 nm sample at both laser powers because it was possible to both observe a signal and avoid damage at both input powers for this particular sample. We also measured the input laser powers used in both regimes using a power meter to be 45 μW (blue) and 112 μW (red). These powers differ by a factor of 2.5 while the factor between the two THG measurements was 18.19. Since a third harmonic signal should scale as the cube of the laser power (and we have proven this to be true for this particular sample in figure 4c), we would expect the signal to have scaled by a factor of 2.5^3 = 15.625, whereas we measured a factor of 18.19. Given that this difference amounts to less than a 3 μW measurement error on the lower power measurement alone (which is within the measurement error of our power meter) and given that the power meter is most certainly the source of this error, we can normalize the THG signal from the smaller gap size samples (blue data points) by the measured factor of 18.19. By doing this we are able to observe the full trend of the experimental data for THG signal vs gap size.

Radiated Third Harmonic Generation Compared to the Electric Field in the Gap:

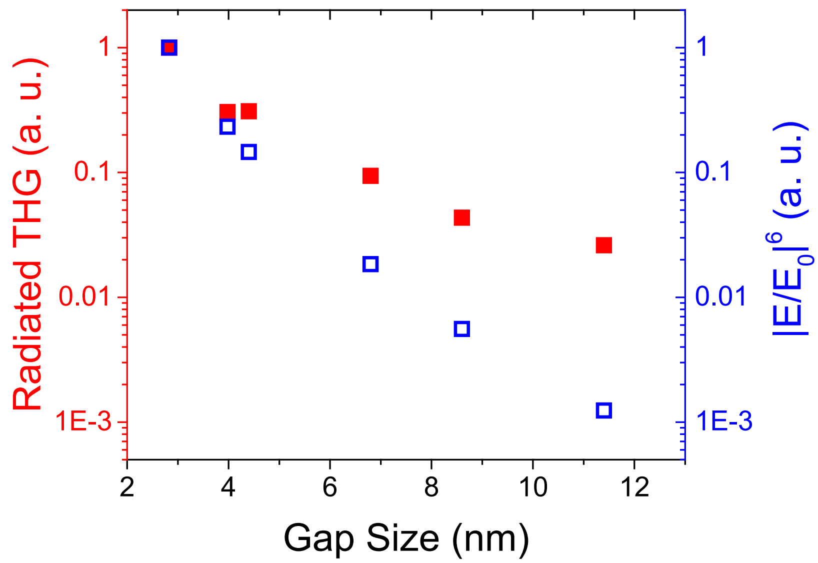

Figure S2: A comparison of the third harmonic generation to the enhanced electric field in the gap. The red solid squares represent the simulated THG radiated away from the stripe sample (same data as the red data points in figure 4a), while the blue hollow squares represent the enhanced electric field in the gap to the sixth power ($|E/E_0|^6$). Because the interest is in the rate of change of these values across the samples, the data points have been normalized by dividing by the max value so that they can be seen on the same scale.

In order to rule out whether the wider stripe widths on the larger gap size samples somehow prevent the THG from coupling out into free space, we have plotted the simulated radiated THG (red solid squares, same as red points in figure 4a) together with the simulated electric field in the gap region to the sixth power, or $|E/E_0|^6$ (blue hollow squares). The THG should scale with $|E/E_0|^6$ since this field is the source of the THG. Here we are interested in only the slope of the trend lines, and thus we have normalized both data sets by dividing by the maximum in order to plot both on the same scale. Both the radiated THG and $|E/E_0|^6$ fall off as the gap size increases (and therefore the stripe width increases). If the larger stripe width is blocking the THG from being radiated, then one would expect that the radiated THG would fall off faster than the electric field in the gap, indicating that some of the THG is not able to escape the gap region. However, the plot above indicates that the radiated THG falls off more slowly than $|E/E_0|^6$, which should indicate that the THG is able to radiate into free space on the samples with larger stripe width. Indeed, the discrepancy between the two data sets suggest that, if anything, the larger stripe samples are more efficient at allowing the THG signal to couple to free space (although a small amount of the discrepancy could also be due to the fact that the alumina layer increases in volume by about a factor of 4 between the smallest and largest gap samples).