Supporting Information

Enhanced Third Harmonic Generation in Ultra-thin Free-standing β-Ga_2O_3 Nanomembranes: Study on Surface and Bulk Contribution

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Derivation of bulk TH field excited at supported and free-standing β-Ga_2O_3

It has been discussed in the main text that a general description of bulk TH field at a sample of thickness l is:

$$E_{Bulk} \propto l * \int_{-z'}^{z'} \frac{\chi(z)dz}{\Delta\epsilon(z) * (1+2iz)^2} \quad (1)$$

where z stands for the coordinate along the optical axis measured from the beam waist position normalized by the confocal parameter. χ is the third-order nonlinear susceptibility at position z. $\Delta\epsilon$ stands for the differences of dielectric constant at fundamental and TH wavelengths at position z. For a supported β-Ga_2O_3 substrate under focused beam as in figure S1, THG is mainly contributed from the focused beam spot area, in our case, beam waist size is around 7.0 μm, THG was found to be strongest when laser focused at the glass/β-Ga_2O_3 interface, consider this interface to be where $z = 0$. The integration can be taken as three parts: from glass, from β-Ga_2O_3 and from air as follows:

$$E_{Bulk} \propto l * \left(\int_{-z'(glass)}^{0} \frac{\chi(glass)dz}{\Delta\epsilon(\text{glass})*(1+2iz)^2} + \int_{0}^{z'(Ga2O3)} \frac{\chi(Ga2O3)dz}{\Delta\epsilon(\text{Ga2O3})*(1+2iz)^2} + \int_{z'(Ga2O3)}^{z'(Air)} \frac{\chi(Air)dz}{\Delta\epsilon(\text{Air})*(1+2iz)^2}\right) \quad (2)$$

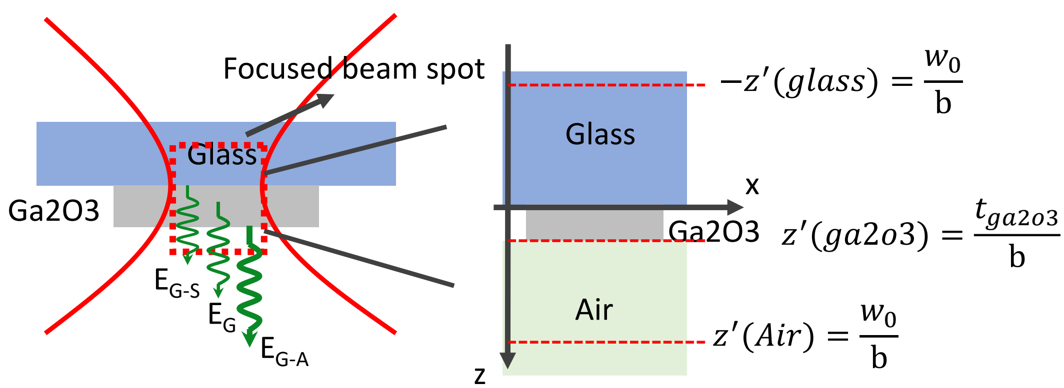

Figure S1. Bulk TH field excited at supported β-Ga_2O_3 sample.

The integration length for glass, β-Ga_2O_3 and air can be determined by $z'(glass) = \frac{w_0}{b}$, $z'(ga2o3) = \frac{t_{ga2o3}}{b}$ and $z'(Air) = \frac{w_0}{b}$ respectively, where w_0 stands for the beam waist size, which is 7.0 μm in our case and b is the confocal parameter which can be calculated by $b = k_\omega \omega_0^2$, where k_ω and ω_0 are the wave number and frequency of the fundamental laser. t_{ga2o3} is the thickness of β-Ga_2O_3 film, which is 991 nm for sample S1. As a result, the normalized integration length for TH excitation for glass, β-Ga_2O_3 and air can be determined to be 0.0362, 0.00512 and 0.0362, respectively.

The integration for the three parts can then be calculated individually as follows:

$$\int_{-z'(glass)}^{0} \frac{\chi_{Glass}^{(3)} dz}{\Delta\epsilon(glass)*(1+2iz)^2} = \frac{\chi_{Glass}^{(3)}}{\Delta\epsilon(glass)} \int_{-z'(glass)}^{0} \frac{dz}{(1+2iz)^2} = \frac{\chi_{Glass}^{(3)}}{\Delta\epsilon(glass)} * \left(\frac{i}{4*i*0+2} - \frac{i}{4*i*(-0.0362)+2}\right) = (0.036 + 0.0026i) * \frac{\chi_{Glass}^{(3)}}{\epsilon_\omega^{Glass} - \epsilon_{3\omega}^{Glass}} \quad (3)$$

$$\int_{0}^{z'(ga2O3)} \frac{\chi_{Ga2O3}^{(3)} dz}{\Delta\epsilon(ga2o3)*(1+2iz)^2} = \frac{\chi_{Ga2O3}^{(3)}}{\Delta\epsilon(ga2o3)} \int_{0}^{z'(ga2O3)} \frac{dz}{(1+2iz)^2} = \frac{\chi_{Ga2O3}^{(3)}}{\Delta\epsilon(ga2o3)} * \left(\frac{i}{4*i*0.00512+2} - \frac{i}{4*i*0+2}\right) = (0.051 - 5.24*10^{-5} i) * \frac{\chi_{Ga2O3}^{(3)}}{\epsilon_\omega^{Ga2O3} - \epsilon_{3\omega}^{Ga2O3}} \quad (4)$$

$$\int_{z'(ga2O3)}^{z'(Air)} \frac{\chi_{air}^{(3)} dz}{\Delta\epsilon(Air)*(1+2iz)^2} = \frac{\chi(Air)}{\Delta\epsilon(Air)} \int_{z'(ga2O3)}^{z'(Air)} \frac{dz}{(1+2iz)^2} = \frac{\chi_{air}^{(3)}}{\Delta\epsilon(Air)} * \left(\frac{i}{4*i*0.0362+2} - \frac{i}{4*i*0.00512+2}\right) = (0.031 - 0.0025i) * \frac{\chi_{air}^{(3)}}{\epsilon_\omega^{air} - \epsilon_{3\omega}^{air}} \quad (5)$$

The magnitude of the real part is more than 30 times larger than the imaginary part, which makes the imaginary parts negligible, and taking the S1 thickness of 991 nm into equation 1, we can get equation 7 in the main text.

$$E_{Bulk}(supported) \propto 0.991 * \left(\frac{0.036\chi_{Glass}^{(3)}}{\epsilon_\omega^{Glass} - \epsilon_{3\omega}^{Glass}} + \frac{0.0052\chi_{Ga2O3}^{(3)}}{\epsilon_\omega^{Ga2O3} - \epsilon_{3\omega}^{Ga2O3}} + \frac{0.031\chi_{air}^{(3)}}{\epsilon_\omega^{air} - \epsilon_{3\omega}^{air}}\right)$$

By applying same method for free-standing β-Ga_2O_3 sample, equation 8 in the main text can also be obtained.