Supplement.

A.

Single layer (2D-sample) is deposited onto a substrate consisting of SiO_2 layer of thickness *d* followed by a much thicker Si material substrate. The incident light at frequency ω (fundamental) is at normal incidence. Non-conducting 2D material is considered. 1→2 interface: air/SiO_2; 2→3 interface: SiO_2/Si . n_2,n_3 – refractive indices of SiO_2 and Si materials [1,2]. The SH intensity enhancement/reduction factor

$$\xi = \left|(1+r_\omega)^2(1+r_{2\omega})\right|^2$$

$$r_\omega = \frac{r_{12} + r_{23}e^{-2i\varphi} - t_{12}\frac{ik\chi}{2}(1+r_{23}e^{-2i\varphi})}{1 + r_{12}r_{23}e^{-2i\varphi} + t_{12}\frac{ik\chi}{2}(1+r_{23}e^{-2i\varphi})}$$

$$r_{12} = \frac{1-n_2-ik\chi}{1+n_2+ik\chi}$$

$$r_{23} = \frac{n_2-n_3}{n_2+n_3}$$

$$t_{12} = 1 - r_{12}$$

$$\varphi = kn_2d$$

$$k = \frac{\omega}{c}$$

$$P = \varepsilon_0\chi E$$

In the formulae above *P* is a linear sheet polarization, χ–linear susceptibility of the monolayer 2D material (and is on the order of 10^{-10} m), n_2,n_3 are refractive indices for SiO_2 and Si at fundamental frequency. The set of expressions for $r_{2\omega}$ is identical to the ones above. However, n_2,n_3 will correspond to the refractive indices at the second harmonic (2ω) frequency.

B.

The electric field for the incident beam at fundamental frequency is represented by the following expression:

$$E_1 = \frac{E_{01}}{2}\left(e^{i\omega t} + e^{-i\omega t}\right) \qquad (1)$$

B.We used the SI system of units.
The intensity is then:

$$I_1 = \frac{1}{2}\varepsilon_0 c\left|E_{01}\right|^2$$

$$E_{01}(x,y) = E_0 e^{-\frac{x^2+y^2}{w_0^2}} \qquad (2)$$

The second harmonic pulse peak power generated in bulk crystal under tight focusing conditions can be represented by the expression that follows from formula (2.58) under (2.99) (p.3609 ref [3]):

$$P_2 = \frac{8\pi\omega_1^2}{\varepsilon_0 c^3 n_2}\frac{1}{\Theta^2}\left|d_{eff}\right|^2 P_1^2 \qquad (3)$$

n_2 – refractive index at SH frequency, Θ – numerical aperture of the microscope objective
Given the fact that the incident and SH beams are partially reflected off the entrance and exit facets correspondingly

$$P_{2\omega}^B = \frac{8\pi\omega_1^2}{\varepsilon_0 c^3 n_2}\frac{1}{\Theta^2}\left|\chi^{(2)}\right|^2\left(\frac{2}{n_1+1}\right)^4\left(\frac{2}{n_2+1}\right)^2 P_\omega^2$$

$$\cong \frac{8\pi\omega_1^2}{\varepsilon_0 c^3 n}\left(\frac{2}{n+1}\right)^6\frac{1}{\Theta^2}\left|d_{eff}\right|^2 P_1^2 \qquad (4).$$

We have neglected with dispersion in the crystal (i.e. $n_2 \approx n_1 = n$) to arrive to expr (4). Also we used $w_0 \approx \lambda/\pi\Theta$ for high NA objective.

SHG in 2D layer.

Solution for the SH field and the corresponding intensity (I_2) can be obtained by using eq. (12) of Ref [4]. Both beams (SH and the fundamental) have the Gaussian spatial distribution and the field's amplitude can be represented via fundamental pulse peak power using

$$P_2^{2D} = (2)^{-1} n_2 \varepsilon_0 c \zeta^2 \int\left|E_{01}^2(x,y)\right|^2 dxdy \qquad (5)$$

and

$$\left|E_0\right|^2 = \frac{4P_1}{\pi\varepsilon_0 c w_0^2}, \qquad (6)$$

The latter relationship is obtained by integrating (2) over space.

Complex enhancement/reduction factor

$$\xi = \left|(1+r_\omega)^2(1+r_{2\omega})\right|^2 \qquad (7),$$

account for interference effect in the two-layered substrate for both fundamental and SH beams [4,5].

Following the solution for the electric field at SH generated in 2D material [4] and taking into account (5) and (6) the peak power at SH frequency ($P_2^{\,2D}$) is:

$$P_2^{2D} = \frac{2P_1^2 k_1^2 \left|\chi^{(2)}\right|^2 \left|(1+r_\omega)^2 (1+r_{2\omega})\right|^2}{n_1^2 \pi \varepsilon_0 c w_0^2} \qquad (8)$$

Then, the ratio (ρ) of the two peak powers (2D_sheet/bulk) is:

$$\rho = \frac{P_2^{2D}}{P_{2\omega}} = \frac{\zeta^2 n (n+1)^6 \Theta^2 \left|\chi_{2D}^{(2)}\right|^2}{256\pi^2 w_0^2 \left|d_{eff}\right|^2} \qquad (9),$$

and

$$\chi_{2D}^{(2)} = \frac{32\pi c\sqrt{\rho}}{(n+1)^3 \Theta^2 \sqrt{\xi}\sqrt{n\omega}} \left|d_{eff}\right| \qquad (10)$$

which is formula (3) in the manuscript.
It is worth reminding that in eq. (10) $\chi_{2D}^{(2)}$ sheet second order nonlinearity is measured in m^2/V.

References used:

1. I. H. Malitson. Interspecimen comparison of the refractive index of fused silica, J. Opt. Soc. Am. **55**, 1205-1208 (1965)
2. D. E. Aspnes and A. A. Studna. Dielectric functions and optical parameters of Si, Ge, GaP, GaAs, GaSb, InP, InAs, and InSb from 1.5 to 6.0 eV, Phys. Rev. B **27**, 985-1009 (1983)
3. G.D. Boyd, and D.A. Kleinman, “Parametric interaction of focused Gaussian light beams” J. Appl. Phys. **39**, 3597-3639(1968)
4. M. Merano, “Nonlinear Optical Response of a Two-dimensional atomic crystal,” Opt. Lett. **41**,187-190 (2016).
5. M. Merano, “Fresnel coefficients of a two-dimensional atomic crystal,” Phy. Rev. A **93**, 013832 (2016)