Supplemental Document

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OPTICAL PHYSICS

Nonlinear optics in gallium phosphide cavities: simultaneous second and third harmonic generation: supplement

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Supplementary material for "Nonlinear optics in gallium phosphide cavities: simultaneous second and third harmonic generation"

Here we present more detailed derivations of the harmonic output powers and inter-modal coupling factors presented in the main text of this work. The second and third harmonic output powers are derived from a system of nonlinear coupled waveguide-cavity mode amplitude equations that are solved for the steady state. The nonlinear inter-modal coupling factors are derived from a perturbation approach which gives the factors as integrals of the mode field components. The crystal symmetry of GaP and the azimuthal symmetry of the microdisk device allows for further simplification of the coupling factors in a manner that directly incorporates phase matching conditions.

1. COUPLED MODE THEORY

The model used in this experiment consists of a three-level cavity mode system a_k with mode frequencies ω_k (where $k = \{1, 2, 3\}$) coupled to a single waveguide mode s with mode frequency ω_0. Start with the input-output cavity mode equation for each cavity mode:

$$\dot{a}_1 = \left(i\omega_1 - \frac{\kappa_1}{2}\right) a_1 + \sqrt{\frac{\kappa_{1,ex}}{2}} s_+, \tag{S1a}$$

$$\dot{a}_2 = \left(i\omega_2 - \frac{\kappa_2}{2}\right) a_2 + \sqrt{\frac{\kappa_{2,ex}}{2}} s_+, \tag{S1b}$$

$$\dot{a}_3 = \left(i\omega_3 - \frac{\kappa_3}{2}\right) a_3 + \sqrt{\frac{\kappa_{3,ex}}{2}} s_+ \tag{S1c}$$

If the cavity possesses both second-order and third-order optical nonlinearities, then the effect of the nonlinear polarization will be to couple the cavity modes to each other. This coupling leads to harmonic generation processes between the cavity modes. If the cavity modes of our three-level system are approximately harmonic to the first mode such that $\omega_2 \approx 2\omega_1$ and $\omega_3 \approx 3\omega_1$, then the second-order nonlinearity will couple modes 1 and 2 due to SHG, and the third-order nonlinearity will couple modes 1 and 3 due to THG. Additionally, the second-order nonlinearity will cause a simultaneous coupling of all three cavity modes due to SFG. We also assume negligible frequency-shifting effects due to third-order nonlinear processes. The full system of coupled-mode equations is then

$$\dot{a}_1 = \left(i\omega_1 - \frac{\kappa_1}{2}\right) a_1 - i\omega_1\beta_{SHG}a_1^*a_2 - i\omega_1\beta_{CSFG}a_2^*a_3 - i\omega_1\beta_{DTHG}\left(a_1^*\right)^2 a_3 + \sqrt{\frac{\kappa_{1,ex}}{2}} s_+, \tag{S2a}$$

$$\dot{a}_2 = \left(i\omega_2 - \frac{\kappa_2}{2}\right) a_2 - i\omega_2\beta_{CSFG}a_1^*a_3 + i\omega_2\beta_{SHG}^*a_1^2 + \sqrt{\frac{\kappa_{2,ex}}{2}} s_+, \tag{S2b}$$

$$\dot{a}_3 = \left(i\omega_3 - \frac{\kappa_3}{2}\right) a_3 + i\omega_3\beta_{CSFG}^*a_1a_2 + i\omega_3\beta_{DTHG}^*a_1^3 + \sqrt{\frac{\kappa_{3,ex}}{2}} s_+ \tag{S2c}$$

where $\beta_{SHG,CSFG,DTHG}$ are the inter-modal coupling factors due to the SHG, CSFG and DTHG processes. We now assume that $\omega_0 \approx \omega_1$ and independently apply rotating-wave approximations at $\omega_k \sim k\omega_0$ to Eqs. (S2) with $k = 1, 2, 3$. Under the rotating-wave approximation, only terms proportional to $\exp(-ik\omega_0)$ will contribute to a_k. The effect of this approximation will be to remove terms proportional to s_+ from Eqs. (S2b-S2c). For the regime of small $|s|$, we also make an undepleted pump approximation for the first harmonic mode [1]. This approximation assumes that the loss in the first harmonic mode due to the nonlinear up-conversions is negligible compared to $|a_1|$, and therefore that the harmonic generation terms may be be removed from Eq. (S2a). Such an approximation is valid when the input power is small (i.e. with input powers on the order of ~ 1 mW or lower), or when the harmonic conversion efficiencies are low. The set of cavity amplitude equations is then

$$\dot{a}_1 = \left(i\Delta_1 - \frac{\kappa_1}{2}\right) a_1 + \sqrt{\frac{\kappa_{1,ex}}{2}} s_+, \tag{S3a}$$

$$\dot{a}_2 = \left(i\Delta_2 - \frac{\kappa_2}{2}\right) a_2 - i\omega_2 \beta_{SF} a_1^* a_3 + i\omega_2 \beta_{SH}^* a_1^2, \tag{S3b}$$

$$\dot{a}_3 = \left(i\Delta_3 - \frac{\kappa_3}{2}\right) a_3 + i\omega_3 \beta_{SF}^* a_1 a_2 + i\omega_3 \beta_{TH}^* a_1^3 \tag{S3c}$$

where $\Delta_k = \omega_k - k\omega_0$ is the detuning of mode k. We now solve for the steady states of Eqs. S2. With the undepleted pump approximation, Eq. (S3a) is easily solved:

$$a_1 = \frac{\sqrt{\frac{\kappa_{1,ex}}{2}}}{i\Delta_1 - \frac{\kappa_1}{2}} s_+ \tag{S4}$$

Substitute Eq. (S4) into Eq. (S3c) and solve for the steady state:

$$\dot{a}_3 = \left(i\Delta_3 - \frac{\kappa_3}{2}\right) a_3 + i\omega_3 \beta_{CSFG}^* a_2 \frac{\sqrt{\frac{\kappa_{1,ex}}{2}}}{i\Delta_1 - \frac{\kappa_1}{2}} s_+ + i\omega_3 \beta_{DTHG}^* \left(\frac{\sqrt{\frac{\kappa_{1,ex}}{2}}}{i\Delta_1 - \frac{\kappa_1}{2}} s_+\right)^3 = 0 \tag{S5}$$

$$\Rightarrow a_3 = -i\omega_3 \beta_{CSFG}^* a_2 \frac{1}{i\Delta_3 - \frac{\kappa_3}{2}} \frac{\sqrt{\frac{\kappa_{1,ex}}{2}}}{i\Delta_1 - \frac{\kappa_1}{2}} s_+ - i\omega_3 \beta_{DTHG}^* \frac{1}{i\Delta_3 - \frac{\kappa_3}{2}} \left(\frac{\sqrt{\frac{\kappa_{1,ex}}{2}}}{i\Delta_1 - \frac{\kappa_1}{2}} s_+\right)^3 \tag{S6}$$

Substitute Eq. (S4) and Eq. (S6) into Eq. (S3b) and solve for the steady state. Since a_2 is in the frame rotating at frequency $\omega_2 \approx 2\omega_0$, this means that only terms proportional to $(s_+)^2$ will contribute. The term in a_3 proportional to s_+^3 can then be neglected so that:

$$\dot{a}_2 \approx \left(i\Delta_2 - \frac{\kappa_2}{2}\right) a_2 - i\omega_2 \beta_{CSFG} \left(\frac{\sqrt{\frac{\kappa_{1,ex}}{2}}}{-i\Delta_1 - \frac{\kappa_1}{2}} s_+^*\right) \left(-i\omega_3 \beta_{CSFG}^* a_2 \frac{1}{i\Delta_3 - \frac{\kappa_3}{2}} \frac{\sqrt{\frac{\kappa_{1,ex}}{2}}}{i\Delta_1 - \frac{\kappa_1}{2}} s_+\right)$$
$$+ i\omega_2 \beta_{SHG}^* \left(\frac{\sqrt{\frac{\kappa_{1,ex}}{2}}}{i\Delta_1 - \frac{\kappa_1}{2}} s_+\right)^2 = 0 \tag{S7}$$

$$\Rightarrow a_2 = \frac{i\omega_2 \beta_{SHG}^* \left(\frac{\sqrt{\frac{\kappa_{1,ex}}{2}}}{i\Delta_1 - \frac{\kappa_1}{2}} s_+\right)^2}{-\left(i\Delta_2 - \frac{\kappa_2}{2}\right) + \omega_2\omega_3 |\beta_{CSFG}|^2 \frac{\frac{\kappa_{1,ex}}{2}|s_+|^2}{\left(i\Delta_3 - \frac{\kappa_3}{2}\right)\left(\Delta_1^2 + \frac{\kappa_1^2}{4}\right)}} \tag{S8}$$

Substitute Eq. (S8) back into Eq. (S6):

$$a_3 = -\omega_2\omega_3 \beta_{SHG}^* \beta_{CSFG}^* \frac{1}{i\Delta_3 - \frac{\kappa_3}{2}} \frac{\left(\frac{\sqrt{\frac{\kappa_{1,ex}}{2}}}{i\Delta_1 - \frac{\kappa_1}{2}} s_+\right)^3}{-\left(i\Delta_2 - \frac{\kappa_2}{2}\right) + \omega_2\omega_3 |\beta_{CSFG}|^2 \frac{\frac{\kappa_{1,ex}}{2}|s_+|^2}{\left(i\Delta_3 - \frac{\kappa_3}{2}\right)\left(\Delta_1^2 + \frac{\kappa_1^2}{4}\right)}} \tag{S9}$$
$$- i\omega_3 \beta_{DTHG}^* \frac{1}{i\Delta_3 - \frac{\kappa_3}{2}} \left(\frac{\sqrt{\frac{\kappa_{1,ex}}{2}}}{i\Delta_1 - \frac{\kappa_1}{2}} s_+\right)^3$$

The output powers are given by Eq. (5) in the main text:

$$P_1 = \left| s_+ - \sqrt{\frac{\kappa_{1,ex}}{2}} a_1 \right|^2, \tag{S10a}$$

$$P_2 = \left| -\sqrt{\frac{\kappa_{2,ex}}{2}} a_2 \right|^2, \tag{S10b}$$

$$P_3 = \left| -\sqrt{\frac{\kappa_{3,ex}}{2}} a_3 \right|^2 \tag{S10c}$$

The full evaluation of Eqs. (S10) is quite complicated, so several simplifying assumptions are made. First, we assume that the waveguide mode is on resonance with the first harmonic mode, $\omega_0 = \omega_1$, so that $\Delta_1 = 0$. This assumption is valid for input frequencies that lie within the first harmonic cavity resonance. Secondly, we assume that, in the limit of weak inter-modal coupling, the harmonic coupling factors due to different generation processes do not interfere with each other. This means that the absolute squares of the coupling factors are separable, i.e. that $|\beta_{CSFG}\beta_{DTHG}|^2 = |\beta_{CSFG}|^2 + |\beta_{DTHG}|^2$. While interference between the CSFG and DTHG processes cannot in general be neglected [1], such an approximation is valid if one or the other processes dominate in contributions to the third harmonic. Evaluating Eqs. (S10) with Eq. (S4) and Eqs. (S8-S9) with these assumptions gives the output powers at resonance as

$$P_1 = P_{in} - \frac{\kappa_{1,ex}^2}{\kappa_1^2} P_{in} \tag{S11}$$

$$P_2 = \frac{8\omega_2^2 |\beta_{SHG}|^2 \frac{\kappa_{1,ex}^2}{\kappa_1^4} \kappa_{2,ex} P_{in}^2}{\Delta_2^2 + \frac{\kappa_2^2}{4} + 4\omega_2\omega_3 |\beta_{CSFG}|^2 \frac{\kappa_{1,ex}}{\kappa_1^2} \frac{\frac{\kappa_2\kappa_3}{4} - \Delta_2\Delta_3}{\Delta_3^2 + \frac{\kappa_3^2}{4}} P_{in} + 4\omega_2^2\omega_3^2 |\beta_{CSFG}|^4 \frac{\kappa_{1,ex}^2}{\kappa_1^4} \frac{1}{\Delta_3^2 + \frac{\kappa_3^2}{4}} P_{in}^2} \tag{S12}$$

$$\begin{aligned} P_3 = {} & 16\omega_3^2 |\beta_{DTHG}|^2 \frac{\kappa_{1,ex}^3}{\kappa_1^6} \frac{\kappa_{3,ex}}{\Delta_3^2 + \frac{\kappa_3^2}{4}} P_{in}^3 \\ & + \frac{16\omega_2^2\omega_3^2 |\beta_{SHG}|^2 |\beta_{CSFG}|^2 \frac{\kappa_{1,ex}^3}{\kappa_1^6} \frac{\kappa_{3,ex}}{\Delta_3^2 + \frac{\kappa_3^2}{4}} P_{in}^3}{\Delta_2^2 + \frac{\kappa_2^2}{4} + 4\omega_2\omega_3 |\beta_{CSFG}|^2 \frac{\kappa_{1,ex}}{\kappa_1^2} \frac{\frac{\kappa_2\kappa_3}{4} - \Delta_2\Delta_3}{\Delta_3^2 + \frac{\kappa_3^2}{4}} P_{in} + 4\omega_2^2\omega_3^2 |\beta_{CSFG}|^4 \frac{\kappa_{1,ex}^2}{\kappa_1^4} \frac{1}{\Delta_3^2 + \frac{\kappa_3^2}{4}} P_{in}^2} \end{aligned} \tag{S13}$$

where $P_{in} = |s_+|^2$. In order to make Eqs. (S12-S13) more amenable to least-squares fitting of power measurements, we factor the $\left(\Delta_2^2 + \frac{\kappa_2^2}{4}\right)$ terms out of the denominators. These equations can then be reduced to a simple form:

$$P_2 = \frac{\eta_{SHG} P_{in}^2}{1 + \zeta_1 P_{in} + \zeta_2 P_{in}^2} \tag{S14}$$

$$P_3 = \eta_{DTHG} P_{in}^3 + \frac{\eta_{CSFG} P_{in}^3}{1 + \zeta_1 P_{in} + \zeta_2 P_{in}^2} \tag{S15}$$

The terms η are the external efficiency terms of each harmonic generation process, given as

$$\eta_{SHG} = 8\omega_2^2 |\beta_{SHG}|^2 \frac{\kappa_{1,ex}^2}{\kappa_1^4} \frac{\kappa_{2,ex}}{\Delta_2^2 + \frac{\kappa_2^2}{4}}, \tag{S16a}$$

$$\eta_{DTHG} = 16\omega_3^2 |\beta_{DTHG}|^2 \frac{\kappa_{1,ex}^3}{\kappa_1^6} \frac{\kappa_{3,ex}}{\Delta_3^2 + \frac{\kappa_3^2}{4}}, \tag{S16b}$$

$$\eta_{CSFG} = 16\omega_2^2\omega_3^2 |\beta_{SHG}|^2 |\beta_{CSFG}|^2 \frac{\kappa_{1,ex}^3}{\kappa_1^6} \frac{1}{\Delta_2^2 + \frac{\kappa_2^2}{4}} \frac{\kappa_{3,ex}}{\Delta_3^2 + \frac{\kappa_3^2}{4}} \tag{S16c}$$

The terms ζ are saturation parameters for linear and quadratic input power scaling, given as

$$\zeta_1 = 4\omega_2\omega_3|\beta_{CSFG}|^2 \frac{\kappa_{1,ex}}{\kappa_1^2} \frac{\frac{\kappa_2\kappa_3}{4} - \Delta_2\Delta_3}{\left(\Delta_2^2 + \frac{\kappa_2^2}{4}\right)\left(\Delta_3^2 + \frac{\kappa_3^2}{4}\right)}, \tag{S17a}$$

$$\zeta_2 = 4\omega_2^2\omega_3^2|\beta_{CSFG}|^4 \frac{\kappa_{1,ex}^2}{\kappa_1^4} \frac{\left(\Delta_2^2 + \frac{\kappa_2^2}{4}\right)\left(\Delta_3^2 + \frac{\kappa_3^2}{4}\right)}{\left(\Delta_2^2 + \frac{\kappa_2^2}{4}\right)^2\left(\Delta_3^2 + \frac{\kappa_3^2}{4}\right)^2} \tag{S17b}$$

For low input powers P_{in}, the harmonic output power scaling Eqs. (S14-S15) reduce to the simple proportional expressions $P_2 \approx \eta_{SHG} P_{in}^2$ and $P_3 \approx (\eta_{DTHG} + \eta_{CSFG}) P_{in}^3$. These simple quadratic and cubic dependencies on the input power are similar to the power dependencies predicted in the cases of SHG [2] or THG as the only nonlinear processes in a coupled resonator. As P_{in} increases, the CSFG process will begin to deplete the second harmonic mode, causing a saturation effect in the second harmonic power output. Likewise, the portion of the third harmonic power output due to CSFG will saturate as well. The result is a sub-quadratic and sub-cubic power scaling in the second and third harmonic outputs respectively. Setting $\Delta_2 = \Delta_3 = 0$ reduces the terms η and ζ to their forms given in Eqs. (12) in the main text. Notably, at triple resonance the saturation parameters satisfy $(\zeta_1/2)^2 = \zeta_2$. This allows us to define a single saturation parameter $\zeta = \zeta_1/2$, which we use in the main text. Our model in principle allows us to quantitatively estimate efficiency and saturation parameters from our data. However, because we do not know the harmonic mode detunings in our experiment, for the power range measured here our model can not reliably distinguish between the triple resonance from the general case. For simplicity, we use the single saturation parameter ζ in the main text, which we find produces good fits with acceptable uncertainties.

2. NONLINEAR MODE COUPLING FACTORS

The nonlinear coupling factors β_{SHG}, β_{CSFG}, β_{DTHG} are determined by the spatial overlap of the fields of each mode participating in a given process. Following the analysis of Rodriquez et al. [3], we derive an expression for the small change in the frequency of a given microdisk mode due to nonlinear effects. When the perturbed mode frequencies are introduced into Eq. (S1), we will obtain the coupled mode equations in Eq. (S3) with explicit expressions for the coupling factors.

We consider a perturbation of the microdisk's dielectric constant $\delta\varepsilon$ which results in a corresponding perturbation in the microdisk's nonlinear polarization $\delta\mathbf{P} = \delta\varepsilon\mathbf{E}$. The resulting fractional change in the mode frequency is proportional to the fraction of electric field energy in the perturbation [4]:

$$\frac{\delta\omega}{\omega} = -\frac{1}{2}\frac{\int_\varepsilon \mathbf{E}^* \cdot \delta\mathbf{P} d^3\mathbf{x}}{\int \varepsilon|\mathbf{E}|^2 d^3\mathbf{x}}, \tag{S18}$$

where $\mathbf{E}$ is the unperturbed electric field. For a three-mode system, we set $\mathbf{E} = \mathbf{E}_1 \exp(-i\omega_1 t) + \mathbf{E}_2 \exp(-i\omega_2 t) + \mathbf{E}_3 \exp(-i\omega_3 t) + \text{c.c.}$, where $\mathbf{E}_{1,2,3}$ are the spatial field profiles of each mode. The second-order nonlinear polarization takes the form $\delta P_i = \varepsilon\chi^{(2)} E_j E_k$, and the third-order nonlinear polarization takes the form $\delta P_i = \varepsilon\chi^{(3)} E_j E_k E_l$, where $\chi^{(2)}$ and $\chi^{(3)}$ are the second- and third-order nonlinear electric susceptibilities respectively, and $\{i, j, k, l\}$ label the $\hat{x}$, $\hat{y}$ and $\hat{z}$ components of $\mathbf{E}$. The total frequency perturbation for a given mode is then obtained by taking a rotating-wave approximation near the mode frequency. In order to maintain the mode amplitude normalization where $|a_k|^2$ is in units of energy, it is necessary to apply an additional normalization term $\left(\int \varepsilon|\mathbf{E}_k|^2 d^3\mathbf{x}\right)^{1/2}$ for each $\mathbf{E}_k$ in the numerator.

Substituting $\omega_k \to \omega_k + \delta\omega_k$ into Eq. (S1) for each mode $k = 1, 2, 3$, rearranging for terms proportional to a_k, comparing to Eq. (S2), and applying the appropriate normalization terms gives the nonlinear coupling factors:

$$\beta_{SHG} = \frac{1}{4}\frac{\int d^3\mathbf{x} \sum_{ijk} \varepsilon\chi_{ijk}^{(2)} \left[E_{1i}^* E_{2j} E_{1k}^* + E_{1i}^* E_{1j}^* E_{2k}\right]}{\int d^3\mathbf{x}\varepsilon|E_1|^2 \left(\int d^3\mathbf{x}\varepsilon|E_2|^2\right)^{1/2}}, \tag{S19}$$

$$\beta_{CSFG} = \frac{1}{4}\frac{\int d^3\mathbf{x} \sum_{ijk} \varepsilon\chi_{ijk}^{(2)} \left[E_{2i}^* E_{3j} E_{1k}^* + E_{2i}^* E_{1j}^* E_{3k}\right]}{\left(\int d^3\mathbf{x}\varepsilon|E_1|^2\right)^{1/2}\left(\int d^3\mathbf{x}\varepsilon|E_2|^2\right)^{1/2}\left(\int d^3\mathbf{x}\varepsilon|E_3|^2\right)^{1/2}}, \tag{S20}$$

$$\beta_{DTHG} = \frac{3}{8}\frac{\int d^3\mathbf{x}\varepsilon\chi^{(3)}\left(\mathbf{E}_1^*\cdot\mathbf{E}_1^*\right)\left(\mathbf{E}_1^*\cdot\mathbf{E}_3\right)}{\left(\int d^3\mathbf{x}\varepsilon|E_1|^2\right)^{1/2}\left(\int d^3\mathbf{x}\varepsilon|E_3|^2\right)^{3/2}}. \tag{S21}$$

The coupling factors can be further simplified depending on the symmetries of the device, as well as the symmetries of the nonlinear susceptibility tensors. GaP possesses a single independent non-zero second-order susceptibility component $d_{14} = d_{25} = d_{36} = 41$ V/pm, where $d_{ij} = \chi_{ij}^{(2)}/2$. Only the off-axis components will then contribute to the sums in Eqs. (S19-S20). The azimuthal symmetry of the microdisk leads to the use of cylindrical coordinates, which thereby incorporates the azimuthal dependence of the fields into the coupling factor calculations. By applying the appropriate vector component transformations [5], the second-order coupling factors are then

$$\beta_{SHG} = \varepsilon d_{14}\int_0^{2\pi}\left(\beta_{SH}^{+}e^{i(2m_1-m_2+2)\varphi} + \beta_{SH}^{-}e^{i(2m_1-m_2-2)\varphi}\right)d\varphi, \tag{S22}$$

$$\beta_{CSFG} = \frac{\varepsilon d_{14}}{2}\int_0^{2\pi}\left(\beta_{SF}^{+}e^{i(m_1+m_2-m_3+2)\varphi} + \beta_{SF}^{-}e^{i(m_1+m_2-m_3-2)\varphi}\right)d\varphi, \tag{S23}$$

where

$$\begin{aligned}\beta_{SHG}^{\pm} = {} & \frac{1}{\int d^3\mathbf{x}\varepsilon|E_1|^2\left(\int d^3\mathbf{x}\varepsilon|E_2|^2\right)^{1/2}}\int\Big[\frac{1}{2}\left(E_{1r}E_{1z}(E_{2r}^* + E_{2\varphi}^*)\right.\\ & + E_{1\varphi}(E_{1r}E_{2z}^* + E_{1z}E_{2r}^*)\big) \pm \frac{i}{4}\big((E_{1r}^2 - E_{1\varphi}^2)E_{2z}^*\\ & + 2E_{1z}(E_{1r}E_{2r}^* - E_{1\varphi}E_{2\varphi}^*)\big)\Big]rdrdz,\end{aligned} \tag{S24}$$

$$\begin{aligned}\beta_{CSFG}^{\pm} = {} & \frac{1}{\left(\int d^3\mathbf{x}\varepsilon|E_1|^2\right)^{1/2}\left(\int d^3\mathbf{x}\varepsilon|E_2|^2\right)^{1/2}\left(\int d^3\mathbf{x}\varepsilon|E_3|^2\right)^{1/2}}\times\\ & \int\Big[E_{1z}^*(E_{2r}^*E_{3\varphi} + E_{2\varphi}^*E_{3r}) + E_{2z}^*(E_{1r}^*E_{3\varphi} + E_{1\varphi}^*E_{3r})\\ & + E_{3z}(E_{1r}^*E_{2\varphi}^* + E_{1\varphi}^*E_{2r}^*) \mp i\big(E_{1z}^*(E_{2r}^*E_{3r} - E_{2\varphi}^*E_{3\varphi})\\ & + E_{2z}^*(E_{1r}^*E_{3r} - E_{1\varphi}^*E_{3\varphi}) + E_{z3}(E_{1r}^*E_{2r}^* - E_{1\varphi}^*E_{2\varphi}^*)\big)\Big]rdrdz.\end{aligned} \tag{S25}$$

Depending on the sign satisfied by the phase matching condition, one of the $\beta^{\pm}$ terms will go to zero leaving the other as the only contributing term.

The equivalent coordinate substitutions for the third-order susceptibility tensor are much more complex. Additionally, the third-order susceptibility of GaP possesses four independent nonzero terms, with only the $\chi_{iiii}^{(3)}$ and $\chi_{iijj}^{(3)}$ terms having been measured near the visible range [6]. We therefore make an approximation that the DTHG coupling factor can be treated as the form of a centrosymmetric crystal with an effective third-order susceptibility equal to the largest third-order susceptibility term of GaP, $\chi_{iiii}^{(3)} = 7.4\times10^{-20}\ \mathrm{V^2\,m^{-2}}$. We believe this approximation is appropriate as, since the first and third harmonic microdisk modes are of the same polarization, that the on-axis terms in the $\chi^{(3)}$ polarization should therefore be much larger than the off-axis terms. Due to this neglecting of off-axis terms, however, our calculated values of β_{DTHG} are likely to be undervalued by some small multiplicative factor. The DTHG coupling factor is then

$$\begin{aligned}\beta_{DTHG} = {} & \frac{3}{8}\frac{\varepsilon\chi_{1111}^{(3)}}{\left(\int d^3\vec{\mathbf{x}}\varepsilon|\vec{\mathbf{E}}_1|^2\right)^{3/2}\left(\int d^3\vec{\mathbf{x}}\varepsilon|\vec{\mathbf{E}}_3|^2\right)^{1/2}}\\ & \times\int_0^{2\pi}\int\Big[e^{i(3m_1-m_3)\varphi}\left((E_{1r}^*)^2 + (E_{1\varphi}^*)^2 + (E_{1z}^*)^2\right)\\ & \times\left(E_{1r}^*E_{3r} + E_{1\varphi}^*E_{3\varphi} + E_{1z}^*E_{3z}\right)\Big]rdrdzd\varphi.\end{aligned} \tag{S26}$$

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