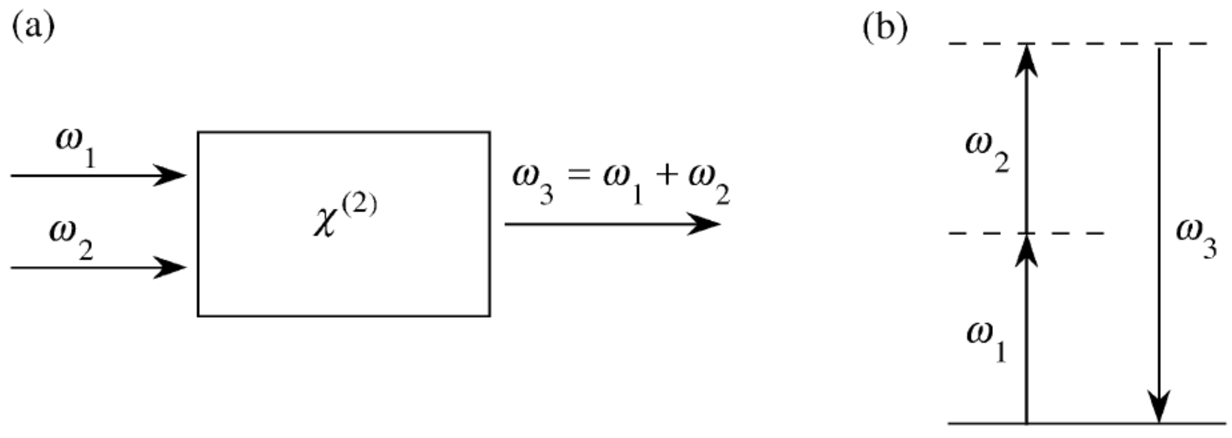
Lecture 6

Sum-frequency and second-harmonic generation.

Sum-Frequency Generation
(frequency up conversion)

Consider sum-frequency generation in a lossless nonlinear optical medium involving collimated, monochromatic, continuous wave input beams.



A photon $\hbar\omega_2$ from the ‘pump’ wave is added to a ‘signal’ photon $\hbar\omega_1$ to geneate an up−converted photon $\hbar\omega_3$

Sum-Frequency Generation

Recall coupled-wave equations for normalized amplitudes *A* for the 3 waves - Lecture 5, eq. (5.9)

$$\frac{dA_1}{dz} = -ig\, A_3 A_2^* e^{-i\Delta kz}$$

$$\frac{dA_2}{dz} = -ig\, A_3 A_1^* e^{-i\Delta kz}$$

$$\frac{dA_3}{dz} = -ig A_1 A_2 e^{i\Delta kz}$$

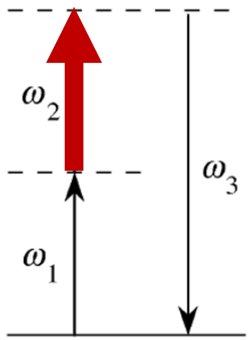
$$A_i = \sqrt{\frac{n_i}{\omega_i}} E_i$$

$$g = \frac{d}{c}\sqrt{\frac{\omega_1\omega_2\omega_3}{n_1 n_2 n_3}}$$ NL coupling coefficient

$$\Delta k = k_3 - k_2 - k_1$$

Sum-Frequency Generation

1) Assume that the **'pump' field at ω_2 is strong** : $E_2 \gg E_1$ and $E_2 \gg E_3$, and we can assume $E_2 \approx$ *const.*
Assume that the 3 waves are phase matched, $\Delta k = 0$ (*k*-vector match), and there is no absorption.



The three equations are then reduced to two:

$$\frac{dA_1}{dz} = -ig\, A_3 A_2^* \qquad (6.1)$$
$$\frac{dA_3}{dz} = -ig A_1 A_2$$

We can also assume (by the proper choice of time origin) A_2 to be real: $A_2 = A_2^*$ (and constant).

define $\boldsymbol{\gamma = gA_2}$ and get:

$$\frac{dA_1}{dz} = -i\gamma\, A_3 \qquad (6.2)$$
$$\frac{dA_3}{dz} = -i\gamma A_1$$

$$\frac{d^2A_1}{dz^2} = -i\gamma(-i\gamma A_1) = -\gamma^2 A_1$$

$$\frac{d^2A_1}{dz^2} + \gamma^2 A_1 = 0$$

likewise $$\frac{d^2A_3}{dz^2} + \gamma^2 A_3 = 0$$

solution: *sines* and *cosines*

Sum-Frequency Generation

For the initial conditions A_1(z=0) =A_{10} and A_3(z=0) =0

we get:

$$A_1 = A_{10}\cos(\gamma z)$$

$$A_3 = -\frac{1}{i\gamma}\frac{dA_1}{dz} = -iA_{10}sin(\gamma z)$$

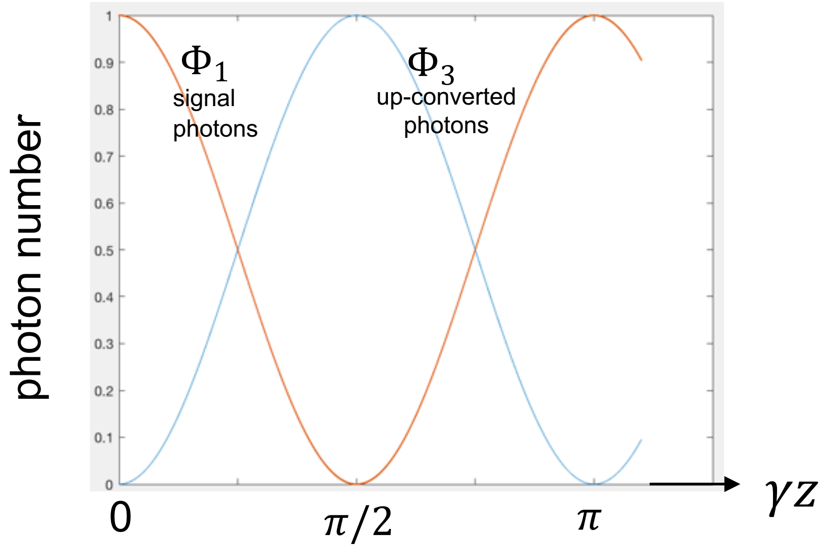
The photon flux densities are

$$\Phi_1 \sim |A_1|^2 = \Phi_{10}\cos^2(\gamma z)$$

$$\Phi_3 \sim |A_3|^2 = \Phi_{10}\sin^2(\gamma z)$$

hence

$$\boxed{\Phi_1 + \Phi_2 = const = \Phi_{10}}$$



The up-conversion efficincy in intensity

$$\eta = \frac{I_3}{I_{10}} = \frac{\omega_3|A_3|^2}{\omega_1|A_{10}|^2} = \frac{\omega_3}{\omega_1}\sin^2(\gamma z)$$

→ max up-conversion efficincy $\eta = \frac{\omega_3}{\omega_1}$ **>1**

Sum-Frequency Generation (low conversion limit)

2) Assume we have **two 'pump' fields at $\boldsymbol{\omega_1}$ and $\boldsymbol{\omega_2}$** and (up) convesion efficiency to ω_3 is small (<< 1 in photons)
Assume that the 3 waves are phase matched, $\Delta k = 0$, and there is no absorption.

The three equations are then reduced to just one: $\frac{dA_3}{dz} = -igA_1A_2$ $\quad A_1, A_2 = const$

solution: $A_3 = -igA_1A_2L$ SFG field grows linearly with distance

L=crystal length

$$|A_3|^2 = g^2 |A_1|^2|A_2|^2L^2$$

$|A|^2 = \frac{I}{\omega}\frac{2}{c\varepsilon_0}$ →

$$\frac{I_3}{\omega_3}\frac{2}{c\varepsilon_0} = g^2 \left(\frac{2}{c\varepsilon_0}\right)^2 \frac{I_1}{\omega_1}\frac{I_2}{\omega_2}L^2$$

$$I_3 = \omega_3{}^2 g^2 \frac{2}{c\varepsilon_0}\frac{I_1I_2}{(\omega_1\omega_2\omega_3)}L^2$$

Finally, for SFG intensity

$$I_3 = \frac{2\omega_3{}^2}{\epsilon_0 c^3}\left(\frac{d^2}{n^3}\right) I_1I_2L^2 \qquad (6.3)$$

nonlinear optical figure of merit

- grows quadratically with distance
- proportional to the product I_1I_2

$$A = \sqrt{\frac{n}{\omega}}E$$

$$I = \frac{1}{2}cn\varepsilon_0|E|^2 = \frac{c\varepsilon_0}{2}\omega|A|^2$$

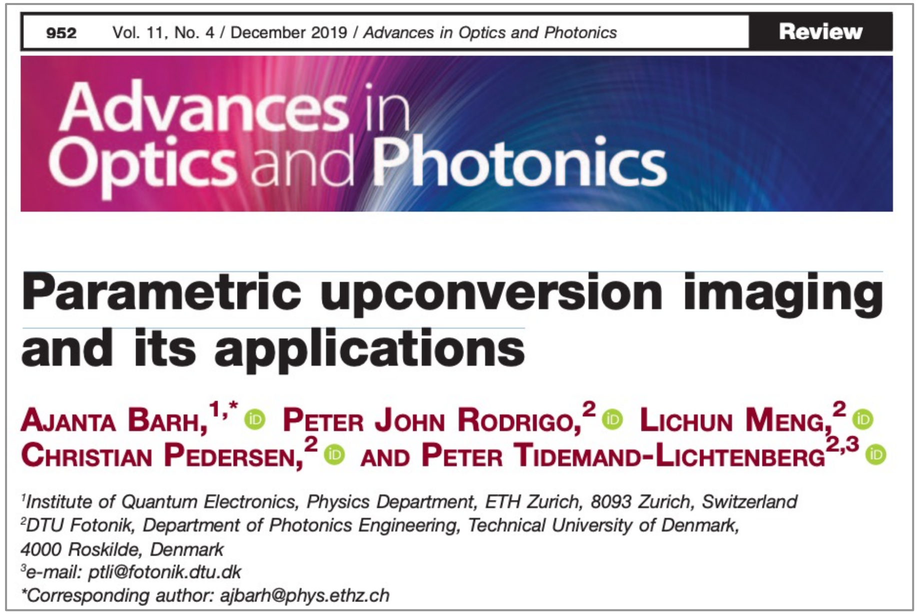
$$g = \frac{d}{c}\sqrt{\frac{\omega_1\omega_2\omega_3}{n_1n_2n_3}}$$

$$g^2 = \frac{d^2}{c^2}\frac{\omega_1\omega_2\omega_3}{n_1n_2n_3} = \frac{\omega_1\omega_2\omega_3}{c^2}\left(\frac{d^2}{\bar{n}^3}\right)$$

$\bar{n} = average\ (n_1, n_2, n_3)$

$d = d_{eff}$ - effective NL coefficient

Detection of mid-infrared light via up-conversion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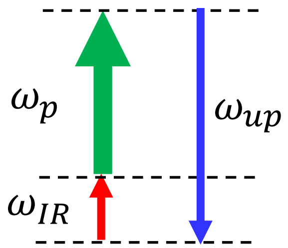
Advances in Optics and Photonics

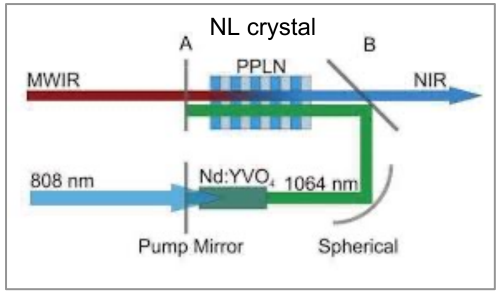
Parametric upconversion imaging and its applications

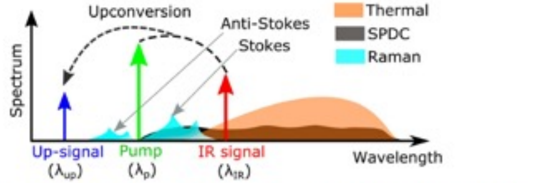
AJANTA BARH,[1,*] PETER JOHN RODRIGO,[2] LICHUN MENG,[2] CHRISTIAN PEDERSEN,[2] AND PETER TIDEMAND-LICHTENBERG[2,3]

[1]*Institute of Quantum Electronics, Physics Department, ETH Zurich, 8093 Zurich, Switzerland*
[2]*DTU Fotonik, Department of Photonics Engineering, Technical University of Denmark, 4000 Roskilde, Denmark*
[3]*e-mail: ptli@fotonik.dtu.dk*
**Corresponding author: ajbarh@phys.ethz.ch*

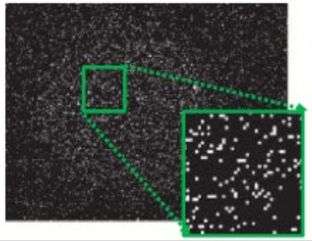


$$\omega_{\mathrm{up}} = \omega_{\mathrm{IR}} + \omega_{\mathrm{p}}.$$





Schematic spectrum of different optical noise sources in a typical short-wavelength-pumped upconversion module. The spectral positions of pump, IR, and upconverted wave are indicated for better understanding. Note that the SPDC and Raman (Stokes and anti-Stokes) photons are generated due to a strong pump.

2007 [44]	[a]**Chemical/spectral upconversion imaging (July 1, 2007)** Imaki and Kobayashi at the University of Fukui, Japan, demonstrated an application of upconversion imaging in chemical sensing (methane detection) at 3.4 μm. Their upconversion detector's sensitivity was 11 times better than a reference direct detector (cooled InAs). Image adapted with permission from [44]. Copyright 2007 Optical Society of America.		Image of methane gas plume
2012 [45]	[a]**Single-photon MIR upconversion imaging (Sept. 16, 2012)** Dam *et al.* at DTU Fotonik, Denmark, first demonstrated upconversion-based single-photon level MIR imaging at room temperature using periodically poled $LiNbO_3$ crystal in a high finesse 1 μm CW laser cavity. The measured dark noise was 0.2 photons/spatial element/s. Figure adapted from [45].		White dots are detected photons

Second Harmonic Generation (SHG)

Second Harmonic Generation (SHG)

Recall (5.2) from the previous lecture: →

$$P(2\omega_1) = \frac{1}{2}\varepsilon_0\chi^{(2)}E_1^2 \quad \text{SHG, second harmonic generation}$$
$$P(2\omega_2) = \frac{1}{2}\varepsilon_0\chi^{(2)}E_2^2 \quad \text{SHG, second harmonic generation}$$
$$P(\omega_1+\omega_2) = \varepsilon_0\chi^{(2)}E_1E_2 \quad \text{SFG, sum-frequency generation} \qquad (5.2)$$
$$P(\omega_1-\omega_2) = \varepsilon_0\chi^{(2)}E_1E_2^* \quad \text{DFG, difference-frequency generation}$$
$$P(0) = \varepsilon_0\chi^{(2)}(E_1E_1^* + E_2E_2^*) \quad \text{OR, optical rectification}$$

The complex amplitude of the nonlinear polarization at $2\omega_1$ is given by :

$$P(2\omega_1) = \frac{1}{2}\varepsilon_0\chi^{(2)}E_1^2 = \varepsilon_0 d_{NL}E_1^2$$

– same for $2\omega_2$

A paradox : why $\frac{1}{2}$ for SHG ?

Regard this example:

$$(\cos\omega_1 t + \cos\omega_2 t)^2 = \cos^2\omega_1 t + \cos^2\omega_2 t + 2\cos\omega_1 t\cdot\cos\omega_2 t =$$

$$= \frac{1}{2}(\cos 2\omega_1 t + 1) + \frac{1}{2}(\cos 2\omega_2 t + 1) + \cos(\omega_1+\omega_2)t + \cos(\omega_1-\omega_2)t =$$

$$= \frac{1}{2}\cos 2\omega_1 t + \frac{1}{2}\cos 2\omega_2 t + \cos(\omega_1+\omega_2)t + \cos(\omega_1-\omega_2)t + 1$$

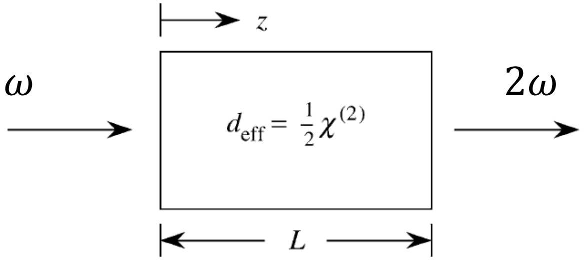
SHG, second harmonic generation | SHG, second harmonic generation | SFG, sum-frequency generation | DFG, difference-frequency generation | OR, optical rectification

Second Harmonic Generation (SHG)

Second Harmonic generation is a degenerate case of 3-wave mixing; we have ω_1=ω_2= ω and ω_3=2ω frequencies

$\omega + \omega = 2\omega$ process $$P_{NL}(2\omega) = \frac{1}{2}\varepsilon_0 \chi^{(2)} E_1^2$$

$2\omega - \omega = \omega$ process $$P_{NL}(\omega) = \varepsilon_0 \chi^{(2)} E_3 E_1^*$$



Coupled-wave equations for normalized amplitudes *A* for the 2 waves

$2\omega - \omega = \omega$ process $$\frac{dA_1}{dz} = -ig\, A_3 A_1^* e^{-i\Delta kz}$$

(6.4)

$\omega + \omega = 2\omega$ process $$\frac{dA_3}{dz} = -i\frac{g}{2} A_1 A_1 e^{i\Delta kz}$$

$$A_i = \sqrt{\frac{n_i}{\omega_i}} E_i$$

$$g = \frac{d}{c}\sqrt{\frac{\omega_1\omega_2\omega_3}{n_1 n_2 n_3}}$$ NL coupling coefficient

$$\Delta k = k_3 - 2k_1$$

Second Harmonic Generation (SHG)

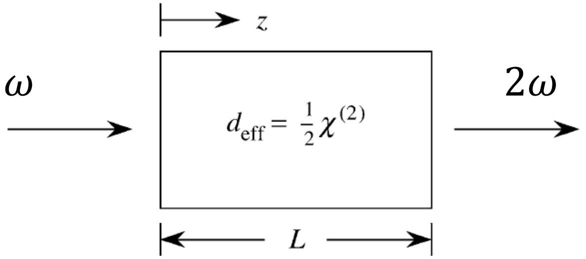
Now let us find how photon fluxes at ω & 2ω are related to each other

photon flux ~ $|A|^2$

$$\frac{d}{dz}|A|^2 = \frac{d}{dz}(AA^*) = A^*\frac{dA}{dz} + A\frac{dA^*}{dz} = A^*\frac{dA}{dz} + c.c.$$

$$A_1^* \quad \frac{dA_1}{dz} = -ig\, A_3 A_1^* e^{-i\Delta kz}$$

$$A_3^* \quad \frac{dA_3}{dz} = -i\frac{g}{2} A_1 A_1 e^{i\Delta kz}$$



$$\frac{d}{dz}|A_1|^2 = -ig\, A_3 A_1^* A_1^* e^{-i\Delta kz} + c.c.$$

$$\frac{d}{dz}|A_3|^2 = -i\frac{g}{2} A_3^* A_1 A_1 e^{i\Delta kz} + c.c. = i\frac{g}{2} A_3 A_1^* A_1^* e^{-i\Delta kz} + c.c.$$

$$\frac{d}{dz}|A_1|^2 + 2\frac{d}{dz}|A_3|^2 = 0 \quad (6.5)$$

$$|A_1(z)|^2 + 2\,|A_3(z)|^2 = A_1(0)^2 \quad (6.6)$$

$$\Phi_1 + 2\Phi_3 = const = \Phi_{10} \quad (6.7)$$

energy conservaion

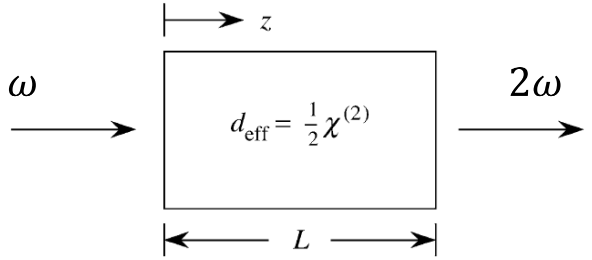
$I = \hbar\omega \cdot \Phi$

$$\frac{d}{dz}(I_\omega + I_{2\omega}) = \frac{d}{dz}(\hbar\omega \cdot \Phi_1 + 2\hbar\omega \cdot \Phi_3) = \hbar\omega\frac{d}{dz}(\Phi_1 + 2\Phi_3) = 0 \quad (6.8)$$

Second Harmonic Generation (SHG)

Now assume that Δk=0 (no phase mismatch), $A_3(z=0)=0$ (no 2ω at the input) and A_1 - real

$$\frac{dA_1}{dz} = -ig\, A_3 A_1^*$$
$$\frac{dA_3}{dz} = -i\frac{g}{2} {A_1}^2 \qquad (6.9)$$



1) Low conversion limit: $A_3 \ll A_1$, $\quad A_1 = const$

$$\frac{dA_3}{dz} = -i\frac{g}{2} {A_1}^2 \approx\ const$$

$$A_3 = -i\frac{g}{2} {A_1}^2 L$$ SH field grows linearly with distance

$$|A_3|^2 = (\frac{g}{2})^2 |A_1|^4 L^2$$

$$|A|^2 = \frac{I}{\omega}\frac{2}{c\varepsilon_0} \longrightarrow \frac{I_3}{2\omega}\frac{2}{c\varepsilon_0} = (\frac{g}{2})^2 (\frac{2}{c\varepsilon_0})^2 (\frac{I_1}{\omega})^2 L^2$$

$$I_3 = {\omega_3}^2\, g^2 \frac{2}{c\varepsilon_0}\frac{I_1 I_2}{\omega_1\omega_2\omega_3} L^2 \qquad I_3 = 4\omega^2 (\frac{g}{2})^2 \frac{2}{c\varepsilon_0}\frac{I_1^2}{(2\omega\cdot\omega\cdot\omega)} L^2$$

$$A = \sqrt{\frac{n}{\omega}} E$$

$$I = \frac{1}{2} cn\varepsilon_0 |E|^2 = \frac{c\varepsilon_0}{2}\omega |A|^2$$

$$\text{g} = \frac{d}{c}\sqrt{\frac{\omega_1\omega_2\omega_3}{n_1 n_2 n_3}}$$

$$g^2 = \frac{d^2}{c^2}\frac{\omega_1\omega_2\omega_3}{n_1 n_2 n_3} = \frac{\omega_1\omega_2\omega_3}{c^2}(\frac{d^2}{\bar{n}^3})$$

$\bar{n} = average\ (n_1, n_2, n_3)$

Finally, SH intensity ($I_3 = I_{2\omega}$)

$$I_{2\omega} = \frac{2\omega^2}{\epsilon_0 c^3}(\frac{d^2}{n^3})\, {I_\omega}^2 L^2 \qquad (6.10a)$$

-grows quadratically with distance
-grows quadratically with I_ω

SHG conversion efficiency in the plane-wave limit:

$$\eta_{2\omega} = I_{2\omega}/I_\omega = \frac{2\omega^2}{\epsilon_0 c^3}(\frac{d^2}{n^3}) I_\omega\, L^2 \qquad (6.10b)$$

Second Harmonic Generation (SHG)

2) High conversion limit

$$\frac{dA_1}{dz} = -ig\, A_3 A_1^*$$
$$\frac{dA_3}{dz} = -i\frac{g}{2} {A_1}^2$$

Assume Δk=0, $A_3(z=0)=0$ (no 2ω input) and A_1 - real

use energy conservation relation: $|A_1(z)|^2 + 2\,|A_3(z)|^2 = A_1(0)^2$

to get $\frac{dA_3}{dz} = -i\frac{g}{2}A_1^2 = -i\frac{g}{2}(A_{10}^2 - 2|A_3|^2)$ – diff. equation for A_3

set $y = i\sqrt{2}A_3$, to get $\frac{dy}{dz} = \frac{g}{\sqrt{2}}(A_{10}^2 - y^2)$

the integral on the left side is reduced to the integral $\int \frac{dx}{1-x^2} = \mathrm{atanh}(x)$

$$\frac{dy}{(A_{10}^2 - y^2)} = \frac{g}{\sqrt{2}}dz \quad \rightarrow \text{integrate} \rightarrow \quad \frac{1}{A_{10}} atanh(\frac{y}{A_{10}}) = \frac{gz}{\sqrt{2}}$$

$$\rightarrow \quad y = A_{10} tanh(\frac{gA_{10}z}{\sqrt{2}}) \qquad A_3 = \frac{1}{i\sqrt{2}}A_{10} tanh(\frac{gA_{10}z}{\sqrt{2}})$$

$$|A_3|^2 = \frac{1}{2}|A_{10}|^2 \tanh^2(\gamma' z) \qquad where\ \gamma' = \frac{gA_{10}}{\sqrt{2}}$$

The photon flux density $\Phi_3 = \frac{1}{2}\Phi_{10}\tanh^2(\gamma' z)$

since $\Phi_1 + 2\Phi_3 = const = \Phi_{10}$

we get $\Phi_1 = \Phi_{10} - 2\Phi_3 = \Phi_{10}\,(1 - \tanh^2(\gamma' z)) = \Phi_{10}\,\mathrm{sech}^2(\gamma' z)$

$$g = \frac{d}{c}\sqrt{\frac{\omega_1\omega_2\omega_3}{n_1 n_2 n_3}}$$

$$\Phi = \frac{c\varepsilon_0}{2\hbar}|A|^2$$

$$\tanh x = \frac{\sinh x}{\cosh x} = \frac{e^x - e^{-x}}{e^x + e^{-x}}$$

$$\mathrm{sech}\, x = \frac{1}{\cosh x} = \frac{2}{e^x + e^{-x}}$$

$$\mathrm{sech}^2 + \tanh^2 = 1$$

Second Harmonic Generation (SHG)

rewrite again:

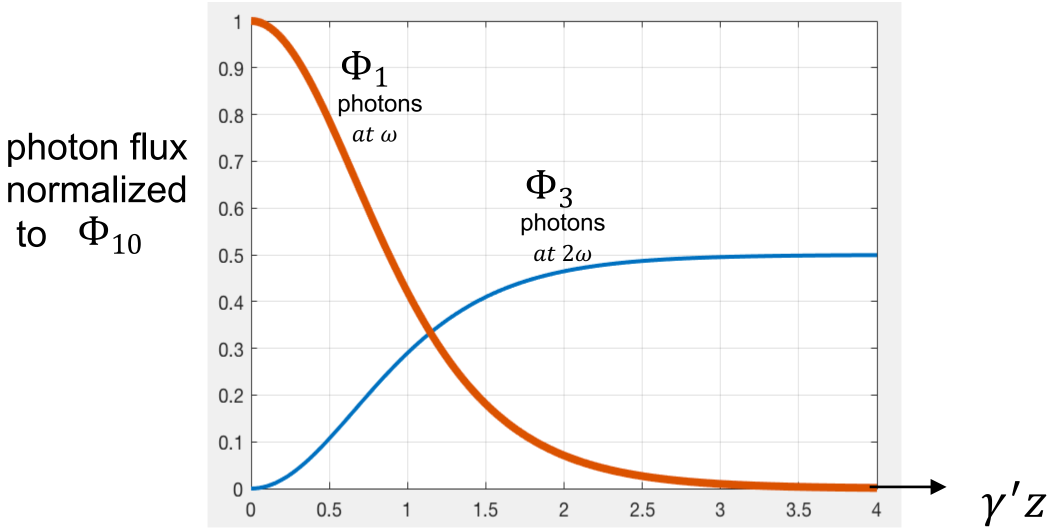
$(\omega)\qquad \Phi_1 = \Phi_{10}\,\text{sech}^2(\gamma' z) \qquad (6.11)$

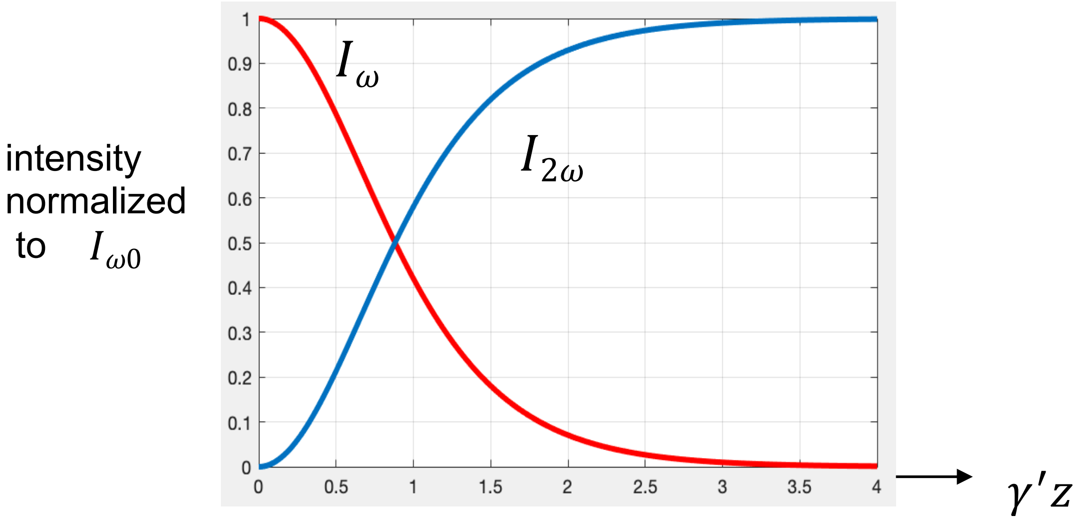
$(2\omega)\qquad \Phi_3 = \frac{1}{2}\Phi_{10}\,\tanh^2(\gamma' z) \qquad (6.12)$

$\gamma' = \frac{gA_{10}}{\sqrt{2}}$

$I_\omega \rightarrow \Phi_1\,\omega \qquad \longrightarrow \qquad I_\omega = I_{\omega 0}\,\text{sech}^2(\gamma' z) \qquad (6.13)$

$I_{2\omega} \rightarrow \Phi_3\,2\omega \qquad \longrightarrow \qquad I_{2\omega} = I_{\omega 0}\tanh^2(\gamma' z) \qquad (6.14)$





Practical example (calculate SHG efficiency)

Problem: calculate the efficiency of SHG conversion from a 1.06 μm laser to the green (0.53 μm)
Laser power: 1W; beam area: (100 μm)^2; nonlinear crystal: L=2 cm; d=20 pm/V; n=2

Use the formula: $$\eta_{2\omega} = I_{2\omega}/I_{\omega} = \frac{2\omega^2}{\epsilon_0 c^3}\left(\frac{d^2}{n^3}\right) I_{\omega}\, L^2 \quad (6.10b)$$

```
P=1; % 1W
lam=1.06e-6;  % 1.06 µm
area=(100e-6)^2; % m^2
L=2e-2; % m
c=3e8; % m/sec
n=2;
eps0=8.85e-12;
d=20e-12; % V/m
omega=2*pi*c/lam;  % rad/sec


eta_SHG=2*omega^2/eps0/c^3 *d^2/n^3 *P/area *L^2
```

=0.053 = **5.3%**