Spontaneous Parametric Down Conversion

We will begin our discussion of SPDC by showing that Maxwell's equations do not predict the spontaneous conversion of a blue photon into two red photons in a nonlinear medium, despite the fact that they do predict the reverse process (second harmonic generation).

Derivation of $\nabla^2 \vec{E} = \frac{1}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}(\chi \vec{E})$:

$\vec{\nabla} \times \vec{E} = -\frac{\partial \vec{B}}{\partial t}$ and $\vec{\nabla} \times \vec{B} = \mu_0 \frac{\partial \vec{D}}{\partial t}$ in a nonmagnetic medium, so we can take the curl of the first eqn:

$$\vec{\nabla} \times (\vec{\nabla} \times \vec{E}) = -\vec{\nabla} \times \frac{\partial \vec{B}}{\partial t} = -\mu_0 \frac{\partial^2 \vec{D}}{\partial t^2}$$

But $\vec{D} \equiv \epsilon_0 (1+\chi)\vec{E}$ where χ is the electric susceptibility, so

$$\vec{\nabla} \times \vec{\nabla} \times \vec{E} = -\mu_0 \epsilon_0 \frac{\partial^2 \vec{E}}{\partial t^2} - \mu_0 \epsilon_0 \frac{\partial^2}{\partial t^2}(\chi \vec{E})$$

Now, maths tell us that $\vec{\nabla} \times (\vec{\nabla} \times \vec{A}) = \vec{\nabla}(\vec{\nabla} \cdot \vec{A}) - \nabla^2 \vec{A}$, so in the absence of stuff like free charges, we have

$$-\nabla^2 \vec{E} = -\mu_0 \epsilon_0 \frac{\partial^2 \vec{E}}{\partial t^2} - \mu_0 \epsilon_0 \frac{\partial^2}{\partial t^2}(\chi \vec{E})$$

Setting $\mu_0 \epsilon_0 \equiv \frac{1}{c_0^2}$, we have

$$\nabla^2 \vec{E} = \frac{1}{c_0^2}\frac{\partial^2 \vec{E}}{\partial t^2} + \frac{1}{c_0^2}\frac{\partial^2 (\chi \vec{E})}{\partial t^2}$$

okay, I've kept that $\chi \vec{E}$ term together for a reason, which is that we will allow for a nonlinear susceptibility. This is a topic that can involve a good bit of vector calulus, but we're just looking for the essence of the effect, so I will assume for the moment that the electric field will always be confined to a particular direction in space.

You may recall the classic definition of the polarization density

$$\vec{P} = \epsilon_0 \chi_e \vec{E} \qquad (P = \epsilon_0 \chi_e E \text{ since we're going 1D})$$

If we allow the electric susceptibility to be nonlinear and 1D, we can write it as a Taylor expansion in E:

$$P = \epsilon_0 \left(\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \cdots \right)$$

$$\equiv P^{(1)} + P^{(2)} + P^{(3)} + \cdots$$

and we have

$$\nabla^2 \vec{E} = \frac{1}{c_0^2} \frac{\partial^2 \vec{E}}{\partial t^2} + \underbrace{\frac{1}{c_0^2} \frac{1}{\epsilon_0}}_{=\mu_0} \frac{\partial^2 \vec{P}^{(1)}}{\partial t^2} + \mu_0 \frac{\partial^2 \vec{P}^{(2)}}{\partial t^2} + \mu_0 \frac{\partial^2 \vec{P}^{(3)}}{\partial t^2} + \cdots$$

[If we identify $\mu = \mu_0$ (non-magnetic medium)]

and $\epsilon^{(1)} \equiv \epsilon_0 (1 + \chi^{(1)})$, we have

$$\nabla^2 \vec{E} = \mu_0 \epsilon_0 (1 + \chi^{(1)}) \frac{\partial^2 \vec{E}}{\partial t^2} + \mu_0 \frac{\partial^2 \vec{P}_{NL}}{\partial t^2}$$

where $\vec{P}_{NL} \equiv \epsilon_0 \chi^{(2)} E^2 + \epsilon_0 \chi^{(3)} E^3 + \cdots$

we can substitute $c^{(1)} \equiv \dfrac{1}{\sqrt{\mu_0 \epsilon^{(1)}}}$ to write

$$\nabla^2 E - \frac{1}{(c^{(1)})^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}_{NL}}{\partial t^2} .$$

The term on the right acts as a "source term" and is a nonlinear function of E, so solving this nonlinear partial differential equation can be challenging. But solving differential equations is all-too-often a laughable process of guess-and-check, so let's explore that technique as applied to a second-order nonlinear medium:

$$|\vec{P}_{NL}| \equiv \epsilon_0 \chi^{(2)} E^2 \quad \text{(with no higher-order terms)}$$

We will guess a solution consisting of three frequencies

$$E(t) = \tfrac{1}{2}\left(\mathcal{E}_1 e^{-i\omega_1 t} + \mathcal{E}_{-1} e^{i\omega_1 t}\right)$$
$$+\tfrac{1}{2}\left(\mathcal{E}_2 e^{-i\omega_2 t} + \mathcal{E}_{-2} e^{i\omega_2 t}\right)$$
$$+\tfrac{1}{2}\left(\mathcal{E}_3 e^{-i\omega_3 t} + \mathcal{E}_{-3} e^{i\omega_3 t}\right)$$

$$\equiv \frac{1}{2}\sum_{q=-3}^{3} \mathcal{E}_q e^{-i\omega_q t}$$ where we define

$$\mathcal{E}_{-q} \equiv \mathcal{E}_q^*$$

$$\omega_{-q} \equiv -\omega_q$$

(and $k_q = \frac{\omega_q}{c^{(1)}}$, though we are absorbing the spatial dependence into $\mathcal{E}_q \sim E_q e^{i\vec{k}_q\cdot\vec{r}}$)

We can write the nonlinear polarization term as

$$P_{NL} = \epsilon_0 \chi^{(2)} E^2$$
$$= \frac{1}{4}\epsilon_0 \chi^{(2)} \sum_{q,r=-3}^{+3} \mathcal{E}_q \mathcal{E}_r e^{-i(\omega_q+\omega_r)t}$$

So the nonlinear source term now looks like

$$\mu_0 \frac{\partial^2 P_{NL}}{\partial t^2} = -\frac{1}{c_0^2}\frac{\chi^{(2)}}{4}\sum_{q,r}(\omega_q+\omega_r)^2 \mathcal{E}_q \mathcal{E}_r e^{-i(\omega_q+\omega_r)t}$$

Plugging this in to our nonlinear wave equation

$$\left(\nabla^2 E - \left(\frac{1}{c^{(1)}}\right)^2 \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_{NL}}{\partial t^2}\right)$$ gives us

$$\frac{1}{2}\sum_q \left(\nabla^2 + k_q^2\right) \mathcal{E}_q e^{-i\omega_q t} = -\frac{1}{4}\frac{\chi^{(2)}}{c_0^2} \sum_{r,s} (\omega_r + \omega_s)^2 \mathcal{E}_r \mathcal{E}_s e^{-i(\omega_r+\omega_s)t}$$

If ω_1, ω_2, and ω_3 are all distinct, we can split up the left side into three terms (one at each frequency) and ask whether the right side can be likewise split up as a sum $\propto -S_1 e^{-i\omega_1 t} - S_2 e^{i\omega_2 t} - S_3 e^{i\omega_3 t}$.

Let's assume it can. We now have

$$(\nabla^2 + k_1^2)\mathcal{E}_1 = -S_1$$

$$(\nabla^2 + k_2^2)\mathcal{E}_2 = -S_2$$

$$(\nabla^2 + k_3^2)\mathcal{E}_3 = -S_3$$

If ω_1, ω_2, and ω_3 are all finite and incommensurate, there is no way to find source term components at frequencies ω_1, ω_2, and ω_3, so that is not a solution.

We will focus on two phenomena associated with the relation

$$\boxed{\omega_1 = \omega_2 \equiv \omega \quad \& \quad \omega_3 \equiv 2\omega}$$

Second-harmonic Generation

We now have two fields to work with, one at frequency (k,ω) and one at $(2k, 2\omega)$. There are now <u>two</u> source terms that oscillate at $+\omega$, and <u>one</u> that oscillates at 2ω:

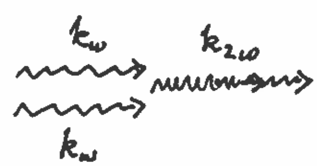
$$\left(\nabla^2 + (k)^2\right)\mathcal{E}_\omega = -\frac{1}{2}\chi^{(2)}\frac{\omega^2}{c^2}\mathcal{E}_{2\omega}\mathcal{E}^*_\omega - \frac{1}{2}\chi^{(2)}\frac{\omega^2}{c^2}\mathcal{E}^*_\omega\mathcal{E}_{2\omega}$$

$$= -\chi^{(2)}\frac{\omega^2}{c^2}\mathcal{E}_{2\omega}\mathcal{E}^*_\omega$$

$$\left(\nabla^2 + (2k)^2\right)\mathcal{E}_{2\omega} = -\frac{1}{2}\chi^{(2)}\frac{(2\omega)^2}{c^2}\mathcal{E}_\omega\mathcal{E}_\omega$$

Now, if we recall that $\mathcal{E}_q = E_q e^{i(\vec{k}_q\cdot\vec{r})}$, it becomes clear that for the source term to stay in phase with the LHS, we require $\vec{k}_{2\omega} = 2\vec{k}_\omega$. So let's simplify this by saying we're going to work with one polarization only and $\vec{k} \parallel \hat{z}$ for all waves. This will accomplish that <u>phase matching condition</u>

$$\boxed{\vec{k}_{2\omega} = 2\vec{k}_\omega}$$



if the nonlinear medium is dispersionless $(\chi^{(1)}_{2\omega} = \chi^{(1)}_\omega)$.

Now, if the amplitudes $E_q(z)$ are slowly-varying wrt z compared to $e^{ik_q z}$, we can approximate

$$\nabla^2 \mathcal{E}_q = \nabla^2 \left(E_q(z) e^{ik_q z} \right)$$

$$= i2k_q e^{ik_q z} \partial_z E_q(z) - k_q^2 E_q(z) e^{ik_q z} + \underbrace{\cancel{e^{ik_q z} \frac{\partial^2 E_q}{\partial z^2}}}_{\text{neglect}}$$

$$\approx e^{ik_q z} \left(i2 k_q \partial_z - k_q^2 \right) E_q(z)$$

$$\therefore \left(\nabla^2 + k_q^2 \right) \mathcal{E}_q \approx e^{ik_q z}\, i2k_q \partial_z E_q(z)$$

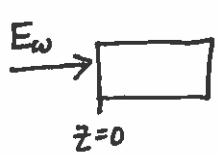
and we derive a set of coupled equations for the slowly-varying amplitudes $E_q(z)$:

$$i2k \frac{dE_\omega}{dz} = -\chi^{(2)} k^2 E_{2\omega} E_\omega^*$$

$$i4k \frac{dE_{2\omega}}{dz} = -2\chi^{(2)} k^2 E_\omega E_\omega$$

$$\Rightarrow \boxed{\begin{aligned} \frac{dE_\omega}{dz} &= \frac{i}{2} \chi^{(2)} k\, E_{2\omega} E_\omega^* \\ \frac{dE_{2\omega}}{dz} &= \frac{i}{2} \chi^{(2)} k\, E_\omega E_\omega \end{aligned}}$$

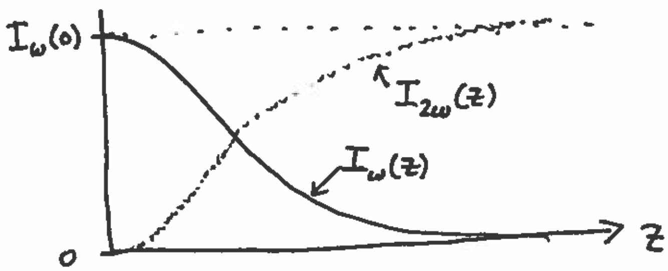
Now let's imagine we're sending "red" light at frequency ω onto a nonlinear crystal that starts at $z=0$:



There is no light at frequency 2ω being input to the system, and we wish to solve for $E_\omega(z)$ and $E_{2\omega}(z)$. The solutions are

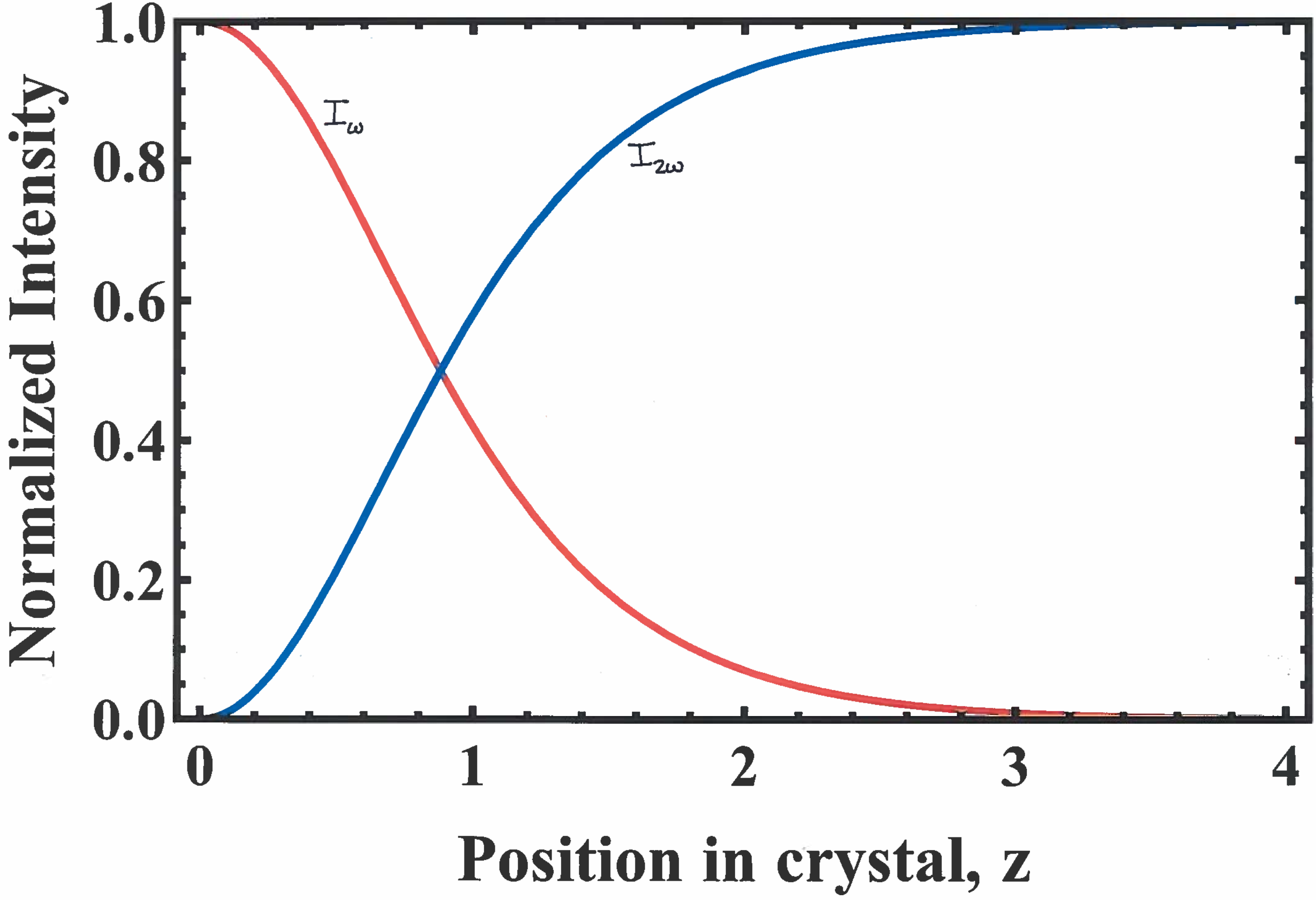
$$\boxed{\begin{aligned} E_\omega(z) &= E_\omega(0)\,\mathrm{sech}\left[\tfrac{1}{2}E_\omega(0)\chi^{(2)}kz\right] \\ E_{2\omega}(z) &= i\,\underset{\text{just }\omega}{E_{\cancel{2\omega}}(0)}\,\tanh\left[\tfrac{1}{2}E_\omega(0)\chi^{(2)}kz\right] \end{aligned}}$$

We can see what is happening by plotting the intensity at ω $\left(I_\omega(z) \equiv \frac{1}{2}c\epsilon_0|E_\omega(z)|^2\right)$ and 2ω as a function of z:

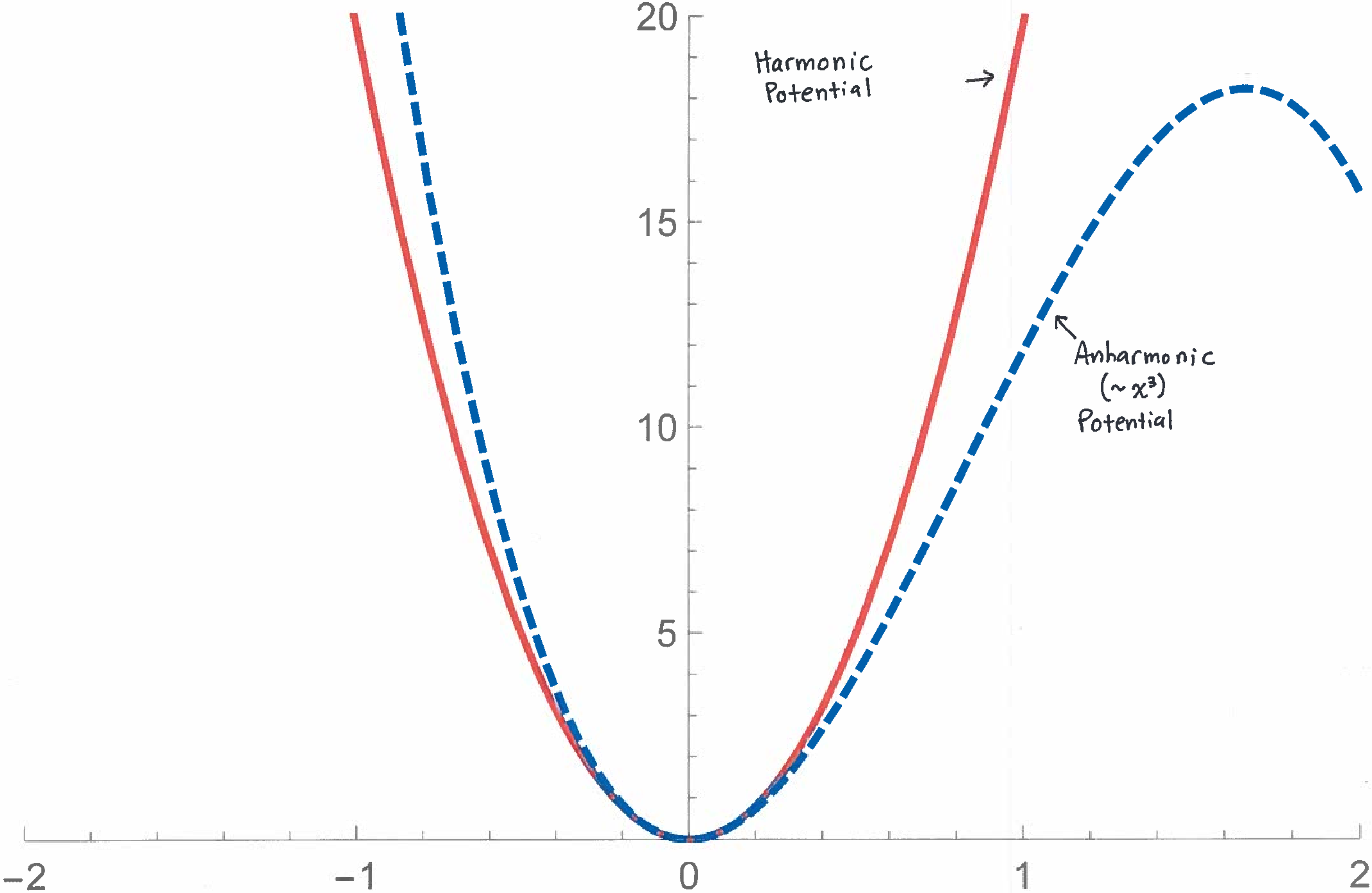


The red light gets weaker and weaker as the blue light gets stronger! Even though there was no blue light incident on the system, the $\chi^{(2)}$ medium is capable ~~to~~ of producing it from red light.

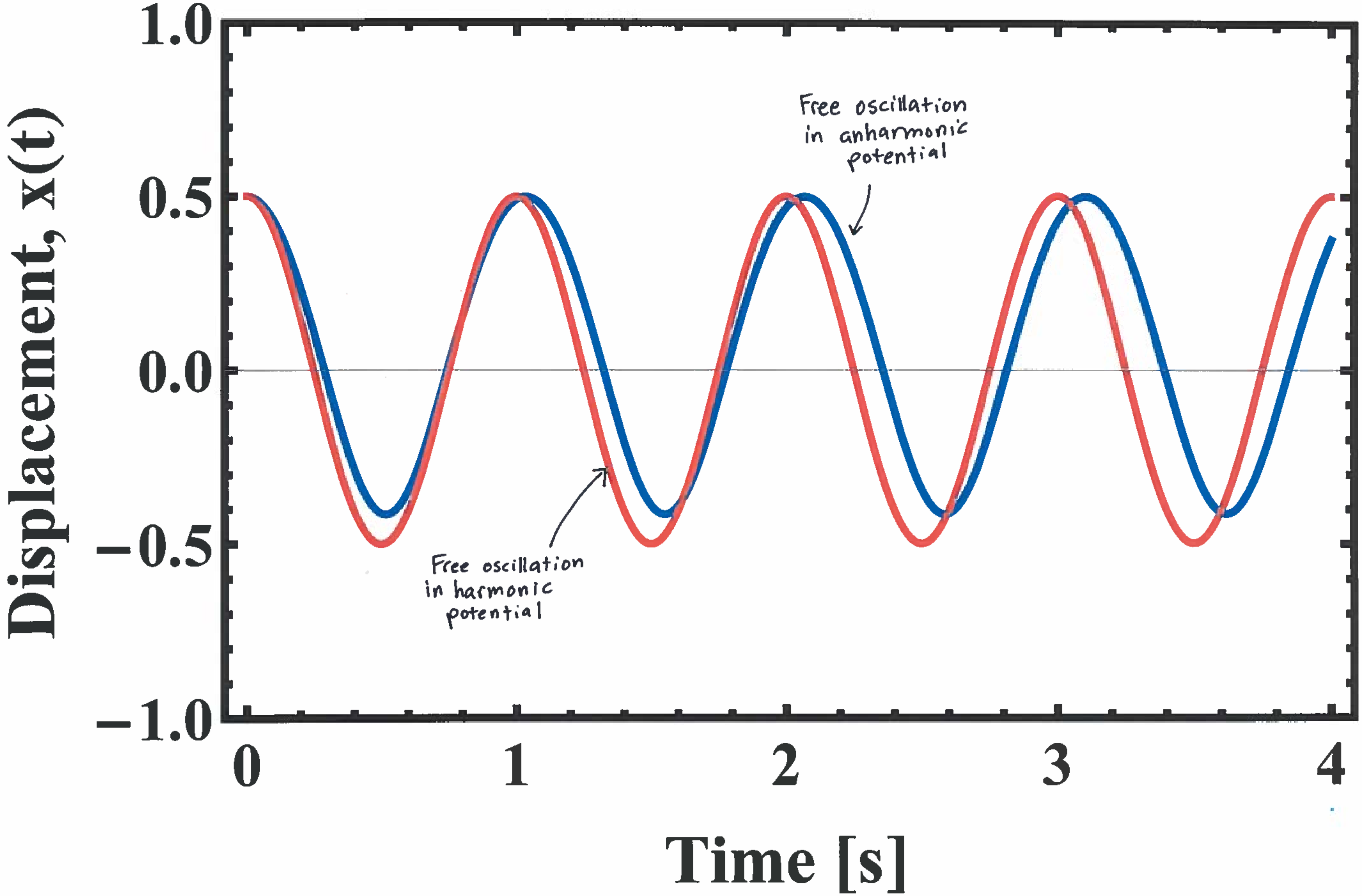
If you want to think about this in terms of photon number, since the blue photons have exactly twice the energy of red photons, we see that <u>pairs</u> of red photons are being converted into blue photons!



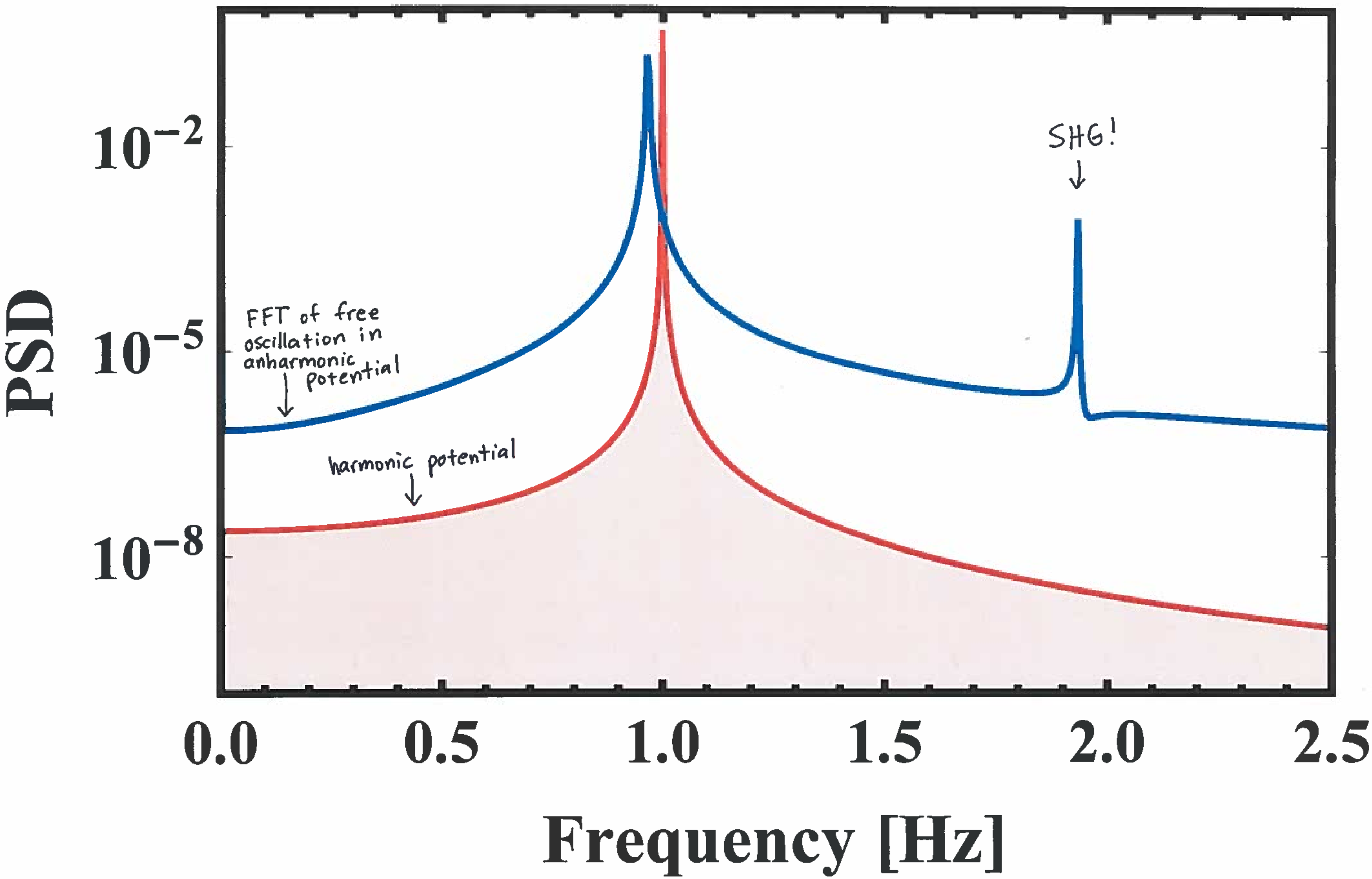
Normalized Intensity
1.0
0.8
0.6
0.4
0.2
0.0
0
1
2
3
4
Position in crystal, z
I_ω
$I_{2\omega}$



20
15
10
5
−2
−1
0
1
2
Harmonic Potential
Anharmonic ($\sim x^3$) Potential



Displacement, x(t)
1.0
0.5
0.0
−0.5
−1.0
0
1
2
3
4
Time [s]
Free oscillation in anharmonic potential
Free oscillation in harmonic potential



PSD
10^{-2}
10^{-5}
10^{-8}
0.0
0.5
1.0
1.5
2.0
2.5
Frequency [Hz]
SHG!
FFT of free oscillation in anharmonic potential
harmonic potential

Spontaneous Parametric Down Conversion

We just saw how classical physics (Maxwell's equations) can be used to describe second harmonic generation (SHG). A beam of red light incident on a $\chi^{(2)}$ crystal can create blue light, even without any quantum mechanics.

But what about the opposite process? Can we send blue light in and make red light? Recall our system of nonlinear equations:

$$\frac{dE_\omega}{dz} = \frac{i}{2}\chi^{(2)} k\, E_{2\omega} E_\omega^*$$

$$\frac{dE_{2\omega}}{dt} = \frac{i}{2}\chi^{(2)} k\, E_\omega E_\omega$$

If $E_{2\omega}$ is zero at $z=0$, there is no source term to nucleate light at ω! So we clearly require some nonzero amplitude for light at frequency ω in order to down-convert one blue photon into two red ones.

Classical physics predicts that there will not be any spontaneous parametric down conversion.