VII. NONLINEAR OPTICS -- CLASSICAL PICTURE:

AN EXTENDED PHENOMENOLOGICAL MODEL OF POLARIZATION:

As an introduction to the subject of nonlinear optical phenomena, we write, in the spirit of Equation [I-4], the most general form of higher order terms in the phenomenological electric field expansion of the polarization density (which may then be inserted in Equations [I-3]) as

$$
\begin{aligned}
\mathbf{P}_\alpha^{(NL)}(\vec{\mathbf{r}},t) = \varepsilon_0 \sum_{\beta\gamma} \int_{\vec{\mathbf{r}}_1}\int_{\vec{\mathbf{r}}_2}\int_{t_1}\int_{t_2} d\vec{\mathbf{r}}_1\, dt_1\, d\vec{\mathbf{r}}_2\, dt_2\, \chi^{(2)}_{\alpha\beta\gamma}\left(\vec{\mathbf{r}}-\vec{\mathbf{r}}_1, t-t_1; \vec{\mathbf{r}}-\vec{\mathbf{r}}_2, t-t_2\right) \mathbf{E}_\beta\left(\vec{\mathbf{r}}_1, t_1\right) \mathbf{E}_\gamma\left(\vec{\mathbf{r}}_2, t_2\right) \\
+\varepsilon_0 \sum_{\beta\gamma\delta} \int_{\vec{\mathbf{r}}_1}\int_{\vec{\mathbf{r}}_2}\int_{\vec{\mathbf{r}}_3}\int_{t_1}\int_{t_2}\int_{t_3} d\vec{\mathbf{r}}_1\, dt_1\, d\vec{\mathbf{r}}_2\, dt_2\, d\vec{\mathbf{r}}_3\, dt_3\, \chi^{(3)}_{\alpha\beta\gamma\delta}\left(\vec{\mathbf{r}}-\vec{\mathbf{r}}_1, t-t_1; \vec{\mathbf{r}}-\vec{\mathbf{r}}_2, t-t_2; \vec{\mathbf{r}}-\vec{\mathbf{r}}_3, t-t_3\right) \\
\times \mathbf{E}_\beta\left(\vec{\mathbf{r}}_1, t_1\right) \mathbf{E}_\gamma\left(\vec{\mathbf{r}}_2, t_2\right) \mathbf{E}_\delta\left(\vec{\mathbf{r}}_3, t_3\right) + \cdots.
\end{aligned}
\qquad [\text{ VII-1 }]
$$

The wave vector and frequency dependent **second and third order susceptibilities** are then defined as

$$
\begin{aligned}
\chi^{(2)}_{\alpha\beta\gamma}\left(\vec{\mathbf{k}}_1, \omega_1; \vec{\mathbf{k}}_2, \omega_2\right) = \int_{\mathbf{R}_1}\int_{\mathbf{R}_2}\int_{\tau_1}\int_{\tau_2} d\vec{\mathbf{R}}_1 d\tau_1\, d\vec{\mathbf{R}}_2 d\tau_2 \exp\left[-i\,\vec{\mathbf{k}}_1\ \vec{\mathbf{R}}_1\right] \exp\left[+i\,\omega_1\,\tau_1\right] \\
\times \exp\left[-i\,\vec{\mathbf{k}}_2\ \vec{\mathbf{R}}_2\right] \exp\left[+i\,\omega_2\,\tau_2\right] \chi^{(2)}_{\alpha\beta\gamma}\left(\vec{\mathbf{R}}_1, \tau_1; \vec{\mathbf{R}}_2, \tau_2\right)
\end{aligned}
\qquad [\text{ VII-2a }]
$$

and

$$
\begin{aligned}
\chi^{(3)}_{\alpha\beta\gamma\delta}\left(\vec{\mathbf{k}}_1, \omega_1; \vec{\mathbf{k}}_2, \omega_2; \vec{\mathbf{k}}_3, \omega_3\right) = \int_{\mathbf{R}_1}\int_{\mathbf{R}_2}\int_{\mathbf{R}_3}\int_{\tau_1}\int_{\tau_2}\int_{\tau_3} d\vec{\mathbf{R}}_1\, d\tau_1\, d\vec{\mathbf{R}}_2\, d\tau_2\, d\vec{\mathbf{R}}_3\, d\tau_3 \exp\left[-i\,\vec{\mathbf{k}}_1\ \vec{\mathbf{R}}_1\right] \exp\left[+i\,\omega_1\,\tau_1\right] \\
\times \exp\left[-i\,\vec{\mathbf{k}}_2\ \vec{\mathbf{R}}_2\right] \exp\left[+i\,\omega_2\,\tau_2\right] \exp\left[-i\,\vec{\mathbf{k}}_3\ \vec{\mathbf{R}}_3\right] \exp\left[+i\,\omega_3\,\tau_3\right] \\
\times \chi^{(3)}_{\alpha\beta\gamma\delta}\left(\vec{\mathbf{R}}_1, \tau_1; \vec{\mathbf{R}}_2, \tau_2; \vec{\mathbf{R}}_3, \tau_3\right)
\end{aligned}
\quad . \qquad [\text{ VII-2b }]
$$

Thus, we may write quite generally [26]

$$
\begin{aligned}
\mathbf{P}_\alpha^{(NL)}(\vec{\mathbf{r}},t) = \varepsilon_0 \sum_{\beta\gamma} \int_{\mathbf{k}_1}\int_{\mathbf{k}_2}\int_{\omega_1}\int_{\omega_2} & d\vec{\mathbf{k}}_1\, d\omega_1\, d\vec{\mathbf{k}}_2\, d\omega_2\, \exp\left[i\left(\vec{\mathbf{k}}_1+\vec{\mathbf{k}}_2\right)\cdot\vec{\mathbf{r}}\right]\exp\left[-i\left(\omega_1+\omega_2\right)t\right] \\
& \times \chi^{(2)}_{\alpha\beta\gamma}\left(\vec{\mathbf{k}}_1,\omega_1;\vec{\mathbf{k}}_2,\omega_2\right)\mathbf{E}_\beta\left(\vec{\mathbf{k}}_1,\omega_1\right)\mathbf{E}_\gamma\left(\vec{\mathbf{k}}_2,\omega_2\right) \\
+\varepsilon_0 \sum_{\beta\gamma\delta} \int_{\mathbf{k}_1}\int_{\mathbf{k}_2}\int_{\mathbf{k}_3}\int_{\omega_1}\int_{\omega_2}\int_{\omega_3} & d\vec{\mathbf{k}}_1\, d\omega_1\, d\vec{\mathbf{k}}_2\, d\omega_2\, d\vec{\mathbf{k}}_3\, d\omega_3\, \exp\left[i\left(\vec{\mathbf{k}}_1+\vec{\mathbf{k}}_2+\vec{\mathbf{k}}_3\right)\cdot\vec{\mathbf{r}}\right]\exp\left[-i\left(\omega_1+\omega_2+\omega_3\right)t\right] \\
& \times \chi^{(3)}_{\alpha\beta\gamma\delta}\left(\vec{\mathbf{k}}_1,\omega_1;\vec{\mathbf{k}}_2,\omega_2;\vec{\mathbf{k}}_3,\omega_3\right)\mathbf{E}_\beta\left(\vec{\mathbf{k}}_1,\omega_1\right)\mathbf{E}_\gamma\left(\vec{\mathbf{k}}_2,\omega_2\right)\mathbf{E}_\delta\left(\vec{\mathbf{k}}_3,\omega_3\right)+\cdots .
\end{aligned}
\qquad [\text{ VII-3 }]
$$

A SIMPLE CLASSICAL MODEL OF NONLINEAR OPTICAL RESPONSE

A simple Lorentz-Dude model is often used in the literature as a valuable guide to the understanding of the frequency behavior of the nonlinear dielectric response.[27] We assume that the potential energy of a one-dimensional nonlinear (anharmonic) oscillator may be written

$$
V(x) = V_2(x)+V_3(x)+V_4(x)+\cdots = \frac{1}{2}M\,\omega_o^2\, x^2+\frac{1}{3}Ma\; x^3+\frac{1}{4}Mb\; x^4+\cdots \qquad [\text{ VII-4 }]
$$

(See figures on next page)

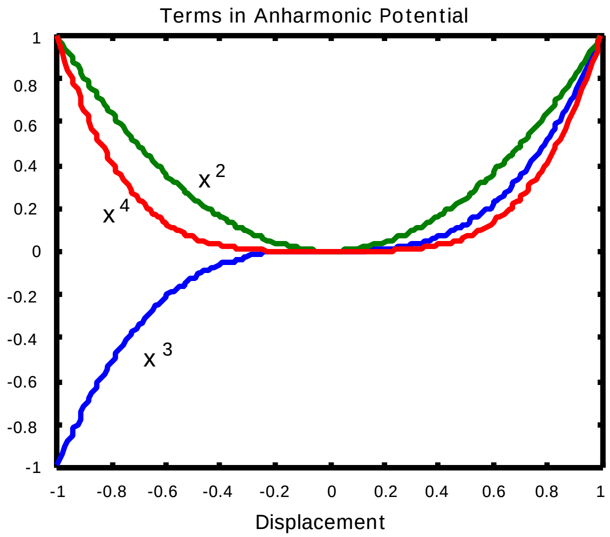
Thus, the equation of motion of a particle moving in that potential becomes

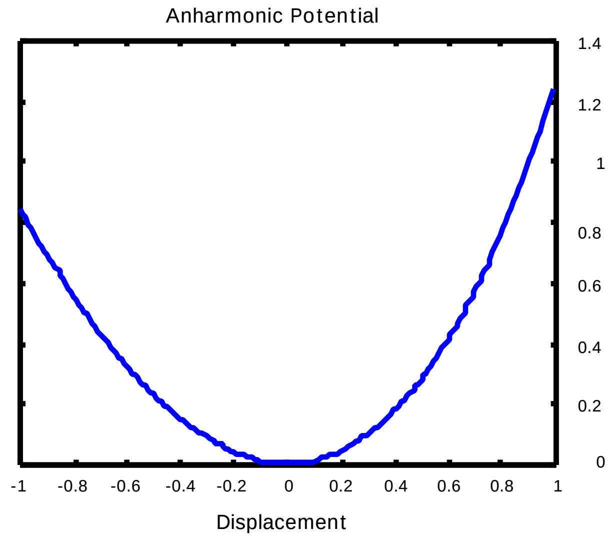
$$
\ddot{x}+\Gamma\,\dot{x}+\omega_o^2\, x+a\,x^2+b\,x^3+\cdots = \frac{q}{M}\,\mathrm{E}\,(t) \qquad [\text{ VII-5 }]
$$

---

26 $\chi^{(2)}_{\alpha\beta\gamma}$ must vanish for any material that is invariant under inversion, since both $\vec{\mathbf{P}}$ and $\vec{\mathbf{E}}$ are vectors, and are thus odd under inversion symmetry. Note also that $\chi^{(3)}_{\alpha\beta\gamma\delta}$ for a given material has the same transformation properties the elastic constants of that material. The nonzero elements of $\chi^{(2)}_{\alpha\beta\gamma}$ and $\chi^{(3)}_{\alpha\beta\gamma\delta}$ for various crystal symmetries are compiled in Y.R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984).

27 See, for example, N. Bloembergen, *Nonlinear Optics* (The Advance Book Program), Addison-Wesley (1992), ISBN 0-201-57868-9.





We can analyze the response of the oscillator by expanding the displacement in powers of the electric field $\mathrm{E}(t)$ -- *viz.*

$$x(t) = x^{(1)}(t) + x^{(2)}(t) + x^{(3)}(t) + \cdots \qquad \text{[VII-6]}$$

where $x^{(n)}(t)$ is proportional to the nth power of the field $\mathrm{E}(t)$. Inserting this expression into Equation [VII-5] and equating like powers of $\mathrm{E}(t)$, we obtain the following hierarchy of equations:

$$\ddot{x}^{(1)}(t) + \Gamma\,\dot{x}^{(1)}(t) + \omega_o^2\,x^{(1)}(t) = (q/M)\,\mathrm{E}(t) \qquad \text{[VII-7a]}$$

$$\ddot{x}^{(2)}(t) + \Gamma\,\dot{x}^{(2)}(t) + \omega_o^2\,x^{(2)}(t) + a\left[x^{(1)}(t)\right]^2 = 0 \qquad \text{[VII-7b]}$$

$$\ddot{x}^{(3)}(t) + \Gamma\,\dot{x}^{(3)}(t) + \omega_o^2\,x^{(3)}(t) + 2a\,x^{(1)}(t)\,x^{(2)}(t) + b\left[x^{(1)}(t)\right]^3 = 0 \qquad \text{[VII-7c]}$$

$$\vdots$$

$$\vdots$$

In the frequency domain we see that

$$-\omega^2 x^{(1)}(\omega) - i\omega\,\Gamma\, x^{(1)}(\omega) + \omega_o^2\, x^{(1)}(\omega) = (q/M)\ \mathrm{E}\,(\omega) \qquad [\text{ VII-8a }]$$

or

$$x^{(1)}(\omega) = (q/M)\ \mathrm{E}\,(\omega)\left[\omega_o^2 - \omega^2 - i\omega\,\Gamma\right]^{-1} = (q/M)\ \mathrm{E}\,(\omega)\ \mathcal{F}(\omega_0, \omega; \Gamma) \qquad [\text{ VII-8b }]$$

where $\mathcal{F}(u, v; w) \equiv \left[u^2 - v^2 - i\,v\ w\right]^{-1}$.[28] Thus, knowing $x^{(1)}(\omega)$, we may then consider $x^{(1)}(t)$ a driving term in Equation [VII-7b] -- *viz*.

$$\ddot{x}^{(2)}(t) + \Gamma\,\dot{x}^{(2)}(t) + \omega_o^2\, x^{(2)}(t) = -a\left[x^{(1)}(t)\right]^2 \qquad [\text{ VII-9 }]$$

Therefore, in the frequency domain

$$x^{(2)}(\omega) = -a\ \mathcal{F}(\omega_0, \omega; \Gamma)\int d\omega'\ x^{(1)}(\omega')\, x^{(1)}(\omega - \omega') \qquad [\text{ VII-10a }]$$

which, in view of Equation [11-8b], becomes

$$\begin{aligned} x^{(2)}(\omega) = -a\,(q/M)^2\ \mathcal{F}[\omega_0, \omega; \Gamma] \\ \times \int d\omega'\ \mathcal{F}[\omega_0, \omega'; \Gamma]\ \mathcal{F}[\omega_0, (\omega - \omega'); \Gamma]\ \mathrm{E}\,(\omega')\ \mathrm{E}\,(\omega - \omega') \end{aligned} \quad .[\text{ VII-10b }]$$

We may treat the third order terms in a similar manner. We write Equation [VII-7c] as

$$\ddot{x}^{(3)}(t) + \Gamma\,\dot{x}^{(3)}(t) + \omega_o^2\, x^{(3)}(t) = -2a\ x^{(1)}(t)\, x^{(2)}(t) - b\left[x^{(1)}(t)\right]^3 \qquad [\text{ VII-11 }]$$

---

28 Note that near resonance

$$\mathcal{F}(\omega_0, \omega; \Gamma) \approx \frac{i}{2\omega}\left[i\,(\omega_0 - \omega) + \Gamma/2\right]^{-1} = \frac{i}{2\omega}\left\{\frac{\Gamma/2 - i\,(\omega_0 - \omega)}{(\omega_0 - \omega)^2 + (\Gamma/2)^2}\right\} = \frac{i}{2\omega}\,\mathcal{D}(\omega_0 - \omega; \Gamma/2)$$

where $\mathcal{D}$ is the so called complex Lorentzian.

In the frequency domain

$$x^{(3)}(\omega) = -2a\ \mathcal{F}(\omega_0,\omega;\Gamma) \int d\omega'\ x^{(1)}(\omega')\ x^{(2)}(\omega-\omega') \\ -\ b\ \mathcal{F}(\omega_0,\omega;\Gamma) \int\int d\omega'\ d\omega''\ x^{(1)}(\omega')\ x^{(1)}(\omega'')\ x^{(1)}(\omega-\omega'-\omega'') \qquad .\quad \text{VII-12]}$$

Using Equations [VII-8b] and [VII-10b], we obtain

$$\begin{aligned} x^{(3)}(\omega) = {} & 2a^2\,(q/M)^3\ \mathcal{F}[\omega_0,\omega;\Gamma] \int\int d\omega'\ d\omega''\ \mathcal{F}[\omega_0,(\omega-\omega');\Gamma]\ \mathcal{F}[\omega_0,\omega';\Gamma] \\ & \times \mathcal{F}[\omega_0,\omega'';\Gamma]\ \mathcal{F}[\omega_0,(\omega'-\omega'');\Gamma]\ \mathrm{E}(\omega-\omega')\ \mathrm{E}(\omega'')\ \mathrm{E}(\omega'-\omega'') \\ & -\, b\,(q/M)^3\ \mathcal{F}[\omega_0,\omega;\Gamma] \int\int d\omega'\ d\omega''\ \mathcal{F}[\omega_0,\omega';\Gamma]\ \mathcal{F}[\omega_0,\omega'';\Gamma] \\ & \times \mathcal{F}[\omega_0,(\omega-\omega'-\omega'');\Gamma]\,\mathrm{E}(\omega')\ \mathrm{E}(\omega'')\ \mathrm{E}(\omega-\omega'-\omega'') \end{aligned} \qquad \text{[VII-13]}$$

In particular, for an input

$$\begin{aligned} \mathrm{E}(t) &= |\mathrm{E}_1|\cos(\omega_1 t+\phi_1) + |\mathrm{E}_2|\cos(\omega_2 t+\phi_2) \\ &= \frac{1}{2}\left[\mathrm{E}_1 \exp(-i\omega_1 t) + \mathrm{E}_1\ \exp(+i\omega_1 t)\right] + \frac{1}{2}\left[\mathrm{E}_2\ \exp(-i\omega_2 t) + \mathrm{E}_2\ \exp(+i\omega_2 t)\right] \end{aligned} \qquad \text{[VII-14]}$$

a. **Second harmonic generation** is due to the terms

$$x^{(2)}(2\omega_1) = -a\,(q/2M)^2\ \mathcal{F}[\omega_0,2\omega_1;\Gamma]\ \mathcal{F}^2[\omega_0,\omega_1;\Gamma]\ \mathrm{E}_1^2(\omega_1) \qquad \text{[VII-15a]}$$

and

$$x^{(2)}(2\omega_2) = -a\,(q/2M)^2\ \mathcal{F}[\omega_0,2\omega_2;\Gamma]\ \mathcal{F}^2[\omega_0,\omega_2;\Gamma]\ \mathrm{E}_2^2(\omega_2) \qquad \text{[VII-15b]}$$

b. **Sum frequency generation** is due to the term

$$x^{(2)}(\omega_1+\omega_2) = -a\,(q/2M)^2\ \mathcal{F}\left[\omega_0,(\omega_1+\omega_2);\Gamma\right] \\ \times\, \mathcal{F}\left[\omega_0,\omega_1;\Gamma\right]\ \mathcal{F}\left[\omega_0,\omega_2;\Gamma\right]\ \mathrm{E}_1(\omega_1)\ \mathrm{E}_2(\omega_2) \qquad [\text{ VII-16a }]$$

c. **Difference frequency generation** is due to the term

$$x^{(2)}(\omega_1-\omega_2) = -a\,(q/2M)^2\ \mathcal{F}\left[\omega_0,(\omega_1-\omega_2);\Gamma\right] \\ \times\, \mathcal{F}\left[\omega_0,\omega_1;\Gamma\right]\ \mathcal{F}\left[\omega_0,\omega_2;\Gamma\right]\ \mathrm{E}_1(\omega_1)\ \mathrm{E}_2(\omega_2) \qquad [\text{ VII-16b }]$$

d. **Optical rectification** or **dc generation** is due to the terms

$$x^{(2)}(0) = -a\,(q/2M)^2\ \mathcal{F}\left[\omega_0,0;\Gamma\right]\left|\mathcal{F}\left[\omega_0,\omega_1;\Gamma\right]\right|^2\left|\mathrm{E}_1(\omega_1)\right|^2 \qquad [\text{ VII-17a }]$$

and

$$x^{(2)}(0) = -a\,(q/2M)^2\ \mathcal{F}\left[\omega_0,0;\Gamma\right]\left|\mathcal{F}\left[\omega_0,\omega_2;\Gamma\right]\right|^2\left|\mathrm{E}_2(\omega_2)\right|^2 \qquad [\text{ VII-17b }]$$

e. **Third harmonic generation** is due to the terms

$$x^{(3)}(3\omega_1) = (q/M)^3\left\{2a^2\ \mathcal{F}\left[\omega_0,2\omega_1;\Gamma\right]-b\right\} \\ \times\, \mathcal{F}\left[\omega_0,3\omega_1;\Gamma\right]\ \mathcal{F}^3\left[\omega_0,\omega_1;\Gamma\right]\mathrm{E}_1^3(\omega_1) \qquad [\text{ VII-18a }]$$

and

$$x^{(3)}(3\omega_2) = (q/M)^3\left\{2a^2\ \mathcal{F}\left[\omega_0,2\omega_2;\Gamma\right]-b\right\} \\ \times\, \mathcal{F}\left[\omega_0,3\omega_2;\Gamma\right]\ \mathcal{F}^3\left[\omega_0,\omega_2;\Gamma\right]\ \mathrm{E}_2^3(\omega_2) \qquad [\text{ VII-18b }]$$

f. **Intensity dependent propagation** is due to the terms

$$x^{(3)}(\omega_1) = (q/2M)^3\left\{2a^2\ \mathcal{F}\left[\omega_0,2\omega_1;\Gamma\right]-b\right\} \\ \times\left|\mathcal{F}\left[\omega_0,\omega_1;\Gamma\right]\right|^2\mathcal{F}^2\left[\omega_0,\omega_1;\Gamma\right]\left|\mathrm{E}_1(\omega_1)\right|^2\mathrm{E}_1(\omega_1) \qquad [\text{ VII-19a }]$$

and

$$x^{(3)}(\omega_2) = (q/2M)^3 \left\{2a^2\ \mathcal{F}[\omega_0, 2\omega_2; \Gamma] - b\right\} \times \left|\mathcal{F}[\omega_0, \omega_2; \Gamma]\right|^2 \mathcal{F}^2[\omega_0, \omega_2; \Gamma]\ \left|E_1(\omega_2)\right|^2 E_1(\omega_2) \qquad \text{[VII-19b]}$$

g. **<u>Raman generation</u>** (inelastic scattering) involves terms like

$$x^{(3)}(2\omega_1 - \omega_2) = 2\,(q/M)^3 \left\{2a^2\ \mathcal{F}[\omega_0, (\omega_1 - \omega_2); \Gamma] + a^2\ \mathcal{F}[\omega_0, 2\omega_1; \Gamma] - b\right\} \times \mathcal{F}[\omega_0, (2\omega_1 - \omega_2); \Gamma]\ \mathcal{F}^2[\omega_0, \omega_1; \Gamma]\ \mathcal{F}\ [\omega_0, \omega_2; \Gamma]\ E_1^2(\omega_1)\ E_2(\omega_2) \qquad \text{[VII-20]}$$

Note that, according to this simple anharmonic oscillator model, Raman generation may be enhanced by a resonance at a frequency $(\omega_1 - \omega_2)$! Also note that, for this model (see Equation [VII-13a] above), the ratio

$$\frac{\chi^{(2)}(\omega)}{\chi^{(1)}(\omega)\ \chi^{(1)}(\omega - \omega_1)\ \chi^{(1)}(\omega_1)} = -a\left(M/N^2 q^3\right) \qquad \text{[VII-21]}$$

is a constant independent of frequency! This observation is consistent with the famous empirical **Miller Rule** which declares that the ratio

$$\Delta_{ijk} = \frac{\chi^{(2)}_{iijk}(\omega_3 = \omega_1 + \omega_2)}{\chi^{(1)}_{ii}(\omega_3)\ \chi^{(1)}_{jj}(\omega_1)\ \chi^{(1)}_{kk}(\omega_2)} \qquad \text{[VII-22]}$$

has only a weak dispersion and is almost a constant for a wide range of materials!

<u>SECOND HARMONIC GENERATION -- PERTURBATION ANALYSIS</u>

We may write the **<u>nonlinear</u>**, macroscopic Maxwell equations in the form

$$\nabla^2\ \vec{\mathbf{E}}(\vec{\mathbf{r}},\omega) - \vec{\nabla}\left[\vec{\nabla}\cdot\ \vec{\mathbf{E}}(\vec{\mathbf{r}},\omega)\right] + \left(\omega^2/c^2\varepsilon_0\right)\bar{\bar{\varepsilon}}\ \ \vec{\mathbf{E}}(\vec{\mathbf{r}},\omega) = -\mu_0\,\omega^2\ \vec{\mathbf{P}}^{(\mathrm{NL})}(\vec{\mathbf{r}},\omega) \qquad \text{[VII-23]}$$

$$\vec{\nabla}\cdot\left[\overline{\overline{\varepsilon}}\cdot\vec{\mathbf{E}}(\vec{\mathbf{r}},\omega)\right] = -\vec{\nabla}\cdot\vec{\mathbf{P}}^{(\mathrm{NL})}(\vec{\mathbf{r}},\omega) \qquad \text{[VII-24]}$$

Suppose that we have an input driving or pump field

$$\vec{\mathbf{E}}(\vec{\mathbf{r}},\omega_1) = \hat{\mathbf{e}}^{(1)}\,\mathrm{E}(\omega_1)\,\exp\left(i\left(k_1 z - \omega_1 t\right)\right) + c.c. \qquad \text{[VII-25]}$$

Then the components of the polarization with frequency $2\omega_1$ are given by

$$\vec{\mathbf{P}}^{(\mathrm{NL})}(z,\omega_2 = 2\omega_1) = \mathrm{E}^2(\omega_1)\;\overline{\overline{\overline{\chi}}}^{(2)}\!:\hat{\mathbf{e}}^{(1)}\,\hat{\mathbf{e}}^{(1)}\,\exp\left[i\left(2k_1 z - 2\omega_1 t\right)\right] + c.c. \qquad \text{[VII- 26]}$$

It is a convenience to resolve this $\mathbf{P}^{(\mathrm{NL})}$ and the resultant second harmonic field into longitudinal and transverse components -- *viz*.

$$\vec{\mathbf{P}}^{(\mathrm{NL})}(z,\omega_2) = \hat{\mathbf{z}}\,\mathrm{P}_{||}^{(\mathrm{NL})}(z,\omega_2) + \vec{\mathbf{P}}_{\perp}^{(\mathrm{NL})}(z,\omega_2)\ . \qquad \text{[VII-27a]}$$

and

$$\vec{\mathbf{E}}(z,\omega_2) = \hat{\mathbf{z}}\,\mathrm{E}_{||}(z,\omega_2) + \vec{\mathbf{E}}_{\perp}(z,\omega_2) \qquad \text{[VII-27b]}$$

Therefore, we may write the ω_2 or second harmonic components of Equations [VII-23] and [VII-24]

$$\begin{aligned} &\frac{\partial^2}{\partial z^2}\vec{\mathbf{E}}_{\perp}(z,\omega_2) + \frac{\omega_2^2}{c^2}\frac{\varepsilon_{\perp}(\omega_2)}{\varepsilon_0}\vec{\mathbf{E}}_{\perp}(z,\omega_2) + \mu_0\,\omega_2^2\,\vec{\mathbf{P}}_{\perp}^{(\mathrm{NL})}(z,\omega_2) \\ &\qquad + \left\{\frac{\omega_2^2}{c^2}\frac{\varepsilon_{||}(\omega_2)}{\varepsilon_0}\mathrm{E}_{||}(z,\omega_2) + \mu_0\,\omega_2^2\,\mathrm{P}_{||}^{(\mathrm{NL})}(z,\omega_2)\right\}\hat{\mathbf{z}} = 0 \end{aligned} \qquad \text{[VII-28a]}$$

and

$$\frac{\partial}{\partial z}\left\{\varepsilon_{||}(\omega_2)\,\mathrm{E}_{||}(z,\omega_2) + \mathrm{P}_{||}^{(\mathrm{NL})}(z,\omega_2)\right\} = 0 \qquad \text{[VII-28b]}$$

To satisfy these equations two conditions must hold -- *viz*.

$$\mathrm{E}_{\|}(z,\omega_2) = -\frac{1}{\varepsilon_{\|}(\omega_2)}\,\mathrm{P}_{\|}^{(\mathrm{NL})}(z,\omega_2) \qquad \text{[VII-29a]}$$

and

$$\left[\frac{\partial^2}{\partial z^2} + k_2^2\right]\vec{\mathbf{E}}_{\perp}(z,\omega_2) = -\mu_0\,\omega_2^2\,\vec{\mathbf{P}}_{\perp}^{(\mathrm{NL})}(z,\omega_2) \qquad \text{[VII-29b]}$$

where

$$k_2 \equiv \frac{\omega_2}{c}\sqrt{\varepsilon_{\perp}(\omega_2)/\varepsilon_0} = 2\omega_1\sqrt{\mu_0\,\varepsilon_{\perp}(2\omega_1)} \qquad \text{[VII-30]}$$

We now write $\vec{\mathbf{E}}_{\perp}(z,\omega_2) = \hat{\mathbf{e}}^{(2)}\,\mathcal{E}(z,\omega_2)\exp(i\,k_2 z)$ where $\mathcal{E}(z,\omega_2)$ is a slowly varying function of z -- *viz*.

$$\begin{aligned}\frac{\partial^2}{\partial z^2}\vec{\mathbf{E}}_{\perp}(z,\omega_2) &= -\hat{\mathbf{e}}^{(2)}\left\{k_2^2\,\mathcal{E}(z,\omega_2) - 2\,i\,k_2\frac{\partial}{\partial z}\mathcal{E}(z,\omega_2) - \frac{\partial^2}{\partial z^2}\mathcal{E}(z,\omega_2)\right\}\exp(i\,k_2 z)\\ &\approx -\left\{k_2^2\,\mathcal{E}(z,\omega_2) - 2\,i\,k_2\frac{\partial}{\partial z}\mathcal{E}(z,\omega_2)\right\}\exp(i\,k_2 z)\end{aligned} \qquad \text{[VII-31]}$$

so that Equation [VII-29b] may be written

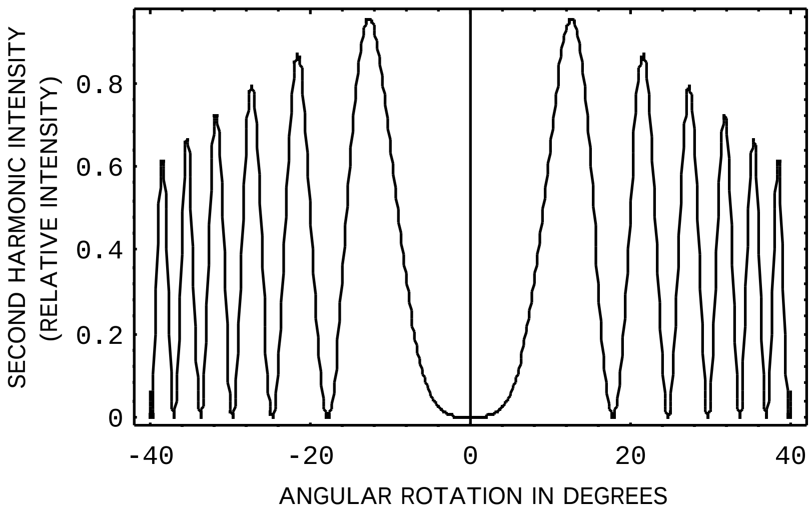
$$\begin{aligned}\frac{\partial}{\partial z}\mathcal{E}(z,\omega_2) &= \frac{i}{2k_2}\mu_0\,\omega_2^2\,\hat{\mathbf{e}}^{(2)}\!:\vec{\mathbf{P}}_{\perp}^{(\mathrm{NL})}(z,\omega_2)\exp(-i\,k_2 z)\\ &= \frac{i\,\mu_0\,\omega_2^2}{2k_2}\,\mathrm{E}^2(\omega_1)\,\hat{\mathbf{e}}^{(2)}\!:\overline{\overline{\overline{\chi}}}^{(2)}\!:\hat{\mathbf{e}}^{(1)}\,\hat{\mathbf{e}}^{(1)}\exp(i\,\Delta k z)\end{aligned} \qquad \text{[VII-32]}$$

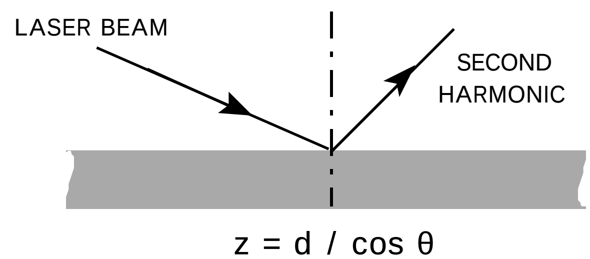
where $\Delta k \equiv 2k_1 - k_2 = \frac{2\omega_1}{c}\left[n_{\mathrm{FH}}(\omega_1) - n_{\mathrm{SH}}(2\omega_1)\right]$.

If we assume that the driving field stays constant, we can directly integrate Equation [VII-32] to obtain the **exceedingly famous and important equation** for the spatial variation of the second harmonic field -- *viz*.

$$\mathcal{E}(z,\omega_2) = z \left\{ \frac{i\,\mu_0\,\omega_2^2}{4\,k_2}\,\hat{\mathbf{e}}^{(2)} : \overline{\overline{\chi}}^{(2)} : \hat{\mathbf{e}}^{(1)}\,\hat{\mathbf{e}}^{(1)}\,\exp\left(i\,\Delta k\,z/2\right) \right\} \mathrm{E}^2(\omega_1)\,\mathrm{sinc}\left[\Delta k z/2\right] . \quad [\text{ VII-33 }]$$

MAKER[29] FRINGES

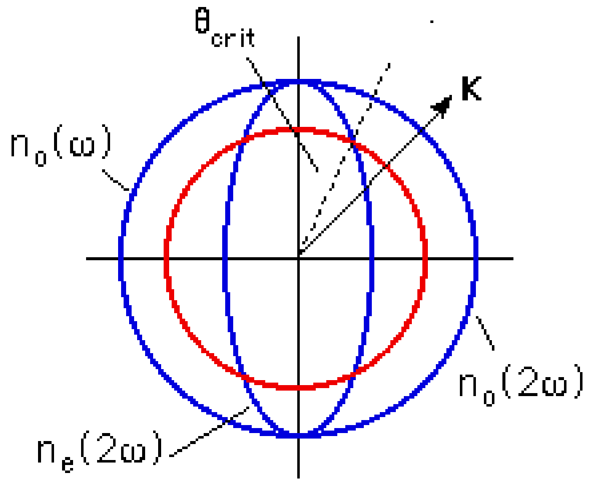


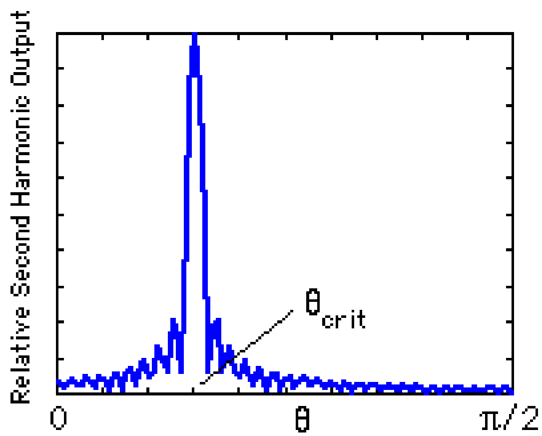


SECOND HARMONIC GENERATION - COUPLED WAVE ANALYSIS

When the process of second harmonic generation takes place under conditions of ***perfect phase match*** -- *i.e.* $k_2 = 2k_1$ -- the perturbation result breaks down if the path is sufficiently long. Under these circumstances the pump beam will be depleted as the second harmonic grows and a solution of Equation [VII-32] must take into account the spatial variation of $\mathrm{E}^2(\omega_1)$. To that end we assume perfect phase matching

[29] P.D. Maker, R.W. Terhune, N. Nisenoff, and C.M. Savage, *Phys. Rev. Lett.*, **8**, 21 (1965).





and rewrite Equation [VII-32] as

$$\frac{\partial}{\partial z}\mathcal{E}(z,\omega_2) = \frac{i\,\mu_0\,\omega_1^2}{k_1}\,\hat{\mathbf{e}}^{(2)}:\vec{\vec{\chi}}^{(2)}:\hat{\mathbf{e}}^{(1)}\,\hat{\mathbf{e}}^{(1)}\,\mathrm{E}^2(z,\omega_1) \qquad \text{[VII-34]}$$

Of course, as the second harmonic grows Equation [VII-3] tells us that a nonlinear polarization at the pump frequency is generated -- *viz*.

$$\vec{\mathbf{P}}^{(NL)}(z,\omega_1) = \mathrm{E}(z,2\omega_1)\,\mathrm{E}\ (z,\omega_1)\left[\vec{\vec{\chi}}^{(2)}:\hat{\mathbf{e}}^{(2)}\,\hat{\mathbf{e}}^{(1)} + \vec{\vec{\chi}}^{(2)}:\hat{\mathbf{e}}^{(1)}\,\hat{\mathbf{e}}^{(2)}\right] \qquad \text{[VII-35]}$$

Repeating the arguments associated with Equations [VII-28] through [VII-31] we may write

$$\frac{\partial}{\partial z}\vec{\mathbf{E}}(z,\omega_1) = \frac{i}{2k_1}\,\mu_0\,\omega_1^2\,\vec{\mathbf{P}}^{(NL)}(z,\omega_1) \qquad \text{[VII-36]}$$

Combining these two equations, we find an equation governing the spatial evolution of $\vec{\mathbf{E}}(z,\omega_1)$ -- *viz.*

$$\frac{\partial}{\partial z}\,\mathrm{E}(z,\omega_1) = \frac{i\,\mu_0\,\omega_1^2}{2k_1}\left[\hat{\mathbf{e}}^{(1)}:\overline{\overline{\overline{\chi}}}^{(2)}:\hat{\mathbf{e}}^{(2)}\;\hat{\mathbf{e}}^{(1)} + \hat{\mathbf{e}}^{(1)}:\overline{\overline{\overline{\chi}}}^{(2)}:\hat{\mathbf{e}}^{(1)}\;\hat{\mathbf{e}}^{(2)}\right]\mathrm{E}(z,2\omega_1)\,\mathrm{E}^{*}(z,\omega_1)\,. \quad [\text{ VII-37 }]$$

Equations [VII-34] and [VII-37] are then the coupled differential equation which describe the coupling of the first and second harmonic fields. Handling all the "vectorness" in these two equation would obscure important issues. Thus, we consider a pair of somewhat less complicated equations which incorporate the essence of the problem -- *viz.*

$$\frac{\partial}{\partial z}\,\mathrm{E}(z,\omega_2) = \frac{i\,\mu_0\,\omega_1^2}{k_1}\,\chi^{(2)}\,\mathrm{E}^2(z,\omega_1) \qquad [\text{ VII-38a }]$$

$$\frac{\partial}{\partial z}\,\mathrm{E}(z,\omega_1) = \frac{i\,\mu_0\,\omega_1^2}{k_1}\,\chi^{(2)}\,\mathrm{E}(z,2\omega_1)\,\mathrm{E}^{*}(z,\omega_1) \qquad [\text{ VII-38b }]$$

To solve these equation we first write $\mathrm{E}(z,\omega_1) = \mathrm{E}(\omega_1)\,\mathrm{f}_1(z)$ and $\mathrm{E}(z,\omega_2) = \mathrm{E}(\omega_1)\,\mathrm{f}_2(z)$ with the boundary conditions $\mathrm{f}_1(0) = 1$ and $\mathrm{f}_2(0) = 0$. Thus, the coupled equations reduce to the dimensionless form

$$\frac{d}{d\xi}\,\mathrm{f}_2(\xi) = i\,\mathrm{f}_1^2(\xi) \qquad [\text{ VII-39a }]$$

and

$$\frac{d}{d\xi}\,\mathrm{f}_1(\xi) = i\,\mathrm{f}_2(\xi)\,\mathrm{f}_1^{*}(\xi) \qquad [\text{ VII-39b }]$$

where $\xi = z/\mathrm{L}_c$ and $\mathrm{L}_c^{-1} = \left[\mu_0\,\omega_1^2\chi^{(2)}\,\mathrm{E}(\omega_1)\right]/k_1$. We next separate $\mathrm{f}_1(\xi)$ and $\mathrm{f}_2(\xi)$ into phase and amplitude parts as

$$\mathrm{f}_{1,2}(\xi) = u_{1,2}(\xi)\,\exp\left[i\,\varphi_{1,2}(\xi)\right] \qquad \text{[VII-40]}$$

and substitute into Equations [VII-39a] and [VII-39b] -- *viz*.

$$\frac{d}{d\xi}u_2(\xi) + i\,u_2(\xi)\,\frac{d}{d\xi}\varphi_2(\xi) = i\,u_1^2(\xi)\,\exp\left\{i\left[2\varphi_1(\xi) - \varphi_2(\xi)\right]\right\} \qquad \text{[VII-41a]}$$

$$\frac{d}{d\xi}u_1(\xi) + i\,u_1(\xi)\,\frac{d}{d\xi}\varphi_1(\xi) = i\,u_1(\xi)\,u_2(\xi)\,\exp\left\{-i\left[2\varphi_1(\xi) - \varphi_2(\xi)\right]\right\} \qquad \text{[VII-41b]}$$

Equating real and imaginary parts of these equations, we find

$$\frac{d}{d\xi}u_2(\xi) = -u_1^2(\xi)\,\sin\left[2\varphi_1(\xi) - \varphi_2(\xi)\right] \qquad \text{[VII-42a]}$$

$$u_2(\xi)\,\frac{d}{d\xi}\varphi_2(\xi) = u_1^2(\xi)\,\cos\left[2\varphi_1(\xi) - \varphi_2(\xi)\right] \qquad \text{[VII-42b]}$$

$$\frac{d}{d\xi}u_1(\xi) = u_1(\xi)\,u_2(\xi)\,\sin\left[2\varphi_1(\xi) - \varphi_2(\xi)\right] \qquad \text{[VII-42c]}$$

$$\frac{d}{d\xi}\varphi_1(\xi) = u_2(\xi)\,\cos\left[2\varphi_1(\xi) - \varphi_2(\xi)\right] \qquad \text{[VII-42d]}$$

Since the phase enters only in the combination $\psi(\xi) = 2\varphi_1(\xi) - \varphi_2(\xi)$ these four equations reduce to three -- *viz*.

$$\frac{d}{d\xi}u_2(\xi) = -u_1^2(\xi)\,\sin\psi(\xi) \qquad \text{[VII-43a]}$$

$$\frac{d}{d\xi}u_1(\xi) = u_1(\xi)\,u_2(\xi)\,\sin\psi(\xi) \qquad \text{[VII-43b]}$$

$$\frac{d}{d\xi}\psi(\xi) = \left[2u_2(\xi) - \frac{u_1^2(\xi)}{u_2(\xi)} \right] \cos\psi(\xi) \qquad \text{[VII-43c]}$$

Combining these three equations, we obtain

$$\tan\psi(\xi)\,\frac{d}{d\xi}\psi(\xi) - 2\frac{1}{u_1(\xi)}\frac{d}{d\xi}u_1(\xi) - \frac{1}{u_2(\xi)}\frac{d}{d\xi}u_2(\xi) = 0 \qquad \text{[VII-44a]}$$

which is, obviously, equivalent to

$$\frac{d}{d\xi}\ln\left[\, u_2(\xi)\, u_1^2(\xi)\, \cos\psi(\xi) \,\right] = 0 \qquad \text{[VII-44b]}$$

or

$$u_2(\xi)\, u_1^2(\xi) \cos\psi(\xi) = \text{const.} \qquad \text{[VII-44c]}$$

Since $u_2(\xi) \to 0$ as $\xi \to 0$ the "const' must be zero and, hence, $\psi(\xi)$ must be $\pi/2$ for all ξ. Thus, the original four coupled equations now reduce to two -- *viz*.

$$\frac{d}{d\xi}u_2(\xi) = u_1^2(\xi) \qquad \text{[VII-45a]}$$

and

$$\frac{d}{d\xi}u_1(\xi) = -u_1(\xi)\, u_2(\xi) \qquad \text{[VII-45b]}$$

Combining these equations, we obtain

$$u_1(\xi)\frac{d}{d\xi}u_1(\xi) + u_2(\xi)\frac{d}{d\xi}u_2(\xi) = \frac{d}{d\xi}\left[u_1^2(\xi) + u_2^2(\xi)\right] = 0 \qquad \text{[VII-46a]}$$

or

$$u_1^2(\xi) + u_2^2(\xi) = 1 \qquad \text{[VII-46b]}$$

which is an assertion of the principle of **energy conservation**. Taking this equation together with Equation [VII-45a] we see that

$$\frac{d\,u_2(\xi)}{1 - u_2^2(\xi)} = d\,\xi \qquad \text{[VII-47a]}$$

or

$$u_2(\xi) = \tanh\xi \qquad \text{[VII-47b]}$$

and

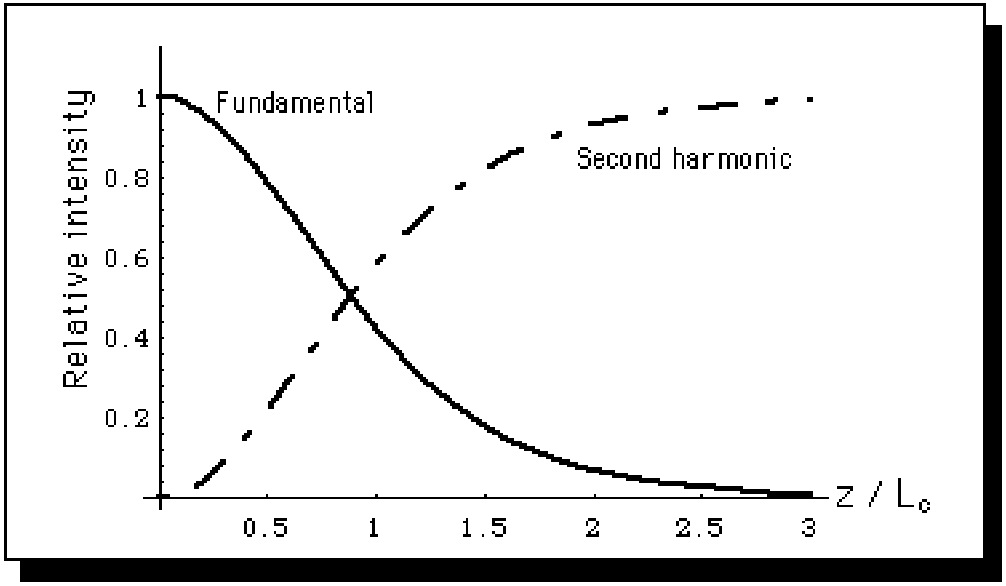
$$d\ln u_1(\xi) = -\tanh(\xi)\; d\,\xi \qquad u_1(\xi) = \frac{1}{\cosh(\xi)} \qquad \text{[VII-47c]}$$

Finally, returning to the original variables, we see that

$$\mathrm{E}(z,\omega_2) = i\,\mathrm{E}(\omega_1)\,\tanh(z/\mathrm{L}_c) \qquad \text{[VII-48a]}$$

and

$$\mathrm{E}(z,\omega_1) = \frac{\mathrm{E}(\omega_1)}{\cosh(z/\mathrm{L}_c)} \qquad \text{[VII-48b]}$$



where $L_c = \dfrac{k_1}{\mu_0 \omega_1^2 \chi^{(2)} E(\omega_1)}$