

1. a)  $F = ma = m\ddot{x} = -\frac{dV}{dx} - eE$

so  $m\ddot{x} + V_0 \frac{x}{a^2} + V_0 \frac{x^2}{a^3} = -eE_0 e^{i\omega t}$

If  $x$  is very small, neglect  $x^2$  term, and

$$\ddot{x} + \frac{V_0}{ma^2} x = -\frac{eE_0}{m} e^{i\omega t}$$

form  $\ddot{x} + \omega_0^2 x = -\frac{eE_0}{m} e^{i\omega t}$

$$\omega_0^2 = \frac{V_0}{ma^2}$$

$$\omega_0 = \sqrt{\frac{V_0}{ma^2}}$$

b) Have  $\ddot{x} = -\omega^2 x_1 e^{i\omega t} - 4\omega^2 x_2 e^{2i\omega t} - 9\omega^2 x_3 e^{3i\omega t}$   
 $\omega_0^2 x = \omega_0^2 x_1 e^{i\omega t} + \omega_0^2 x_2 e^{2i\omega t} + \omega_0^2 x_3 e^{3i\omega t}$   
 $x^2 = x_1^2 e^{2i\omega t} + x_2^2 e^{4i\omega t} + x_3^2 e^{6i\omega t}$   
 $+ 2x_1 x_2 e^{3i\omega t} + 2x_1 x_3 e^{4i\omega t} + 2x_2 x_3 e^{5i\omega t}$

Ignore terms higher than  $e^{3i\omega t}$ , since we don't care about their amplitudes.

$$x^2 \rightarrow x_1^2 e^{2i\omega t} + 2x_1 x_2 e^{3i\omega t}$$

So equation of motion becomes

$$-\omega^2 x_1 e^{i\omega t} - 4\omega^2 x_2 e^{2i\omega t} - 9\omega^2 x_3 e^{3i\omega t}$$

$$+ \omega_0^2 x_1 e^{i\omega t} + \omega_0^2 x_2 e^{2i\omega t} + \omega_0^2 x_3 e^{3i\omega t}$$

$$+ \beta x_1^2 e^{2i\omega t} + 2\beta x_1 x_2 e^{3i\omega t} = -\frac{e}{m} E_0 e^{i\omega t}$$

$$\beta = \frac{V_0}{ma^3} = \frac{\omega_0^2}{a}$$

Coefficients of each  $e^{i\omega t}$  factor are independent, (2)  
 so

$$(1) \quad -\omega^2 x_1 + \omega_0^2 x_1 = -\frac{e}{m} E_0$$

$$(2) \quad -4\omega^2 x_2 + \omega_0^2 x_2 + \beta x_1^2 = 0$$

$$(3) \quad -9\omega^2 x_3 + \omega_0^2 x_3 + 2\beta x_1 x_2 = 0$$

From (1),

$$x_1 = \frac{e}{m} E_0 \frac{1}{\omega^2 - \omega_0^2}$$

From (2),

$$x_2 = \frac{\beta}{4\omega^2 - \omega_0^2} x_1^2 =$$

$$\frac{e^2}{m^2} E_0^2 \frac{\beta}{(4\omega^2 - \omega_0^2)(\omega^2 - \omega_0^2)^2}$$

From (3),

$$x_3 = \frac{2\beta}{9\omega^2 - \omega_0^2} x_1 x_2$$

$$x_3 = \frac{e^3 E_0^3}{m^3} \frac{2\beta^2}{(9\omega^2 - \omega_0^2)(4\omega^2 - \omega_0^2)(\omega^2 - \omega_0^2)^3}$$

c) IF  $\omega \ll \omega_0$

$$x_1 \approx -\frac{e}{m} E_0 \frac{1}{\omega_0^2} = -\frac{e}{m} E_0 \frac{m c^2}{V_0} = -e E_0 \frac{a^2}{V_0}$$

$$x_2 = -\frac{e^2}{m^2} E_0^2 \frac{\beta}{\omega_0^6} = -\frac{e^2}{m^2} E_0^2 \frac{V_0}{m a^3} \frac{a^6 m^3}{V_0^3}$$

$$= -e^2 E_0^2 \frac{a^3}{V_0^2}$$

$$x_3 = \frac{e^3 E_0^3}{m^3} \frac{2\beta^2}{\omega_0^{10}} = 2 \frac{e^3 E_0^3}{m^3} \frac{V_0^2}{m^2 a^6} \frac{m^5 a^{10}}{V_0^5}$$

$$= 2 e^3 E_0^3 \frac{a^4}{V_0^3}$$

So

$$P = -e \times N$$

$$= N e^2 E \frac{a^2}{v_0} + N e^3 E^2 \frac{a^3}{v_0^2} - 2 N e^4 E^3 \frac{a^4}{v_0^3}$$

$$= \chi_0 E + 2d E^2 + 4X^{(3)} E^3$$

$$S_0 \quad \chi = N \frac{e^2 a^2}{\epsilon_0 v_0}$$

$$d = \frac{1}{2} N \frac{e^3 a^3}{v_0^2}$$

$$X^{(3)} = -\frac{1}{2} N \frac{e^4 a^4}{v_0^3}$$

Can write

$$P = N e a \left\{ \frac{e E a}{v_0} + \left( \frac{e E a}{v_0} \right)^2 - 2 \left( \frac{e E a}{v_0} \right)^3 \right\}$$

So if  $e E \ll \frac{v_0}{a}$ , terms are decreasing.

2. We have

$$P(2\omega_1) = d E(\omega_1)^2$$

$$P(2\omega_2) = d E(\omega_2)^2$$

$$P(\omega_1 + \omega_2) = 2d E(\omega_1) E(\omega_2)$$

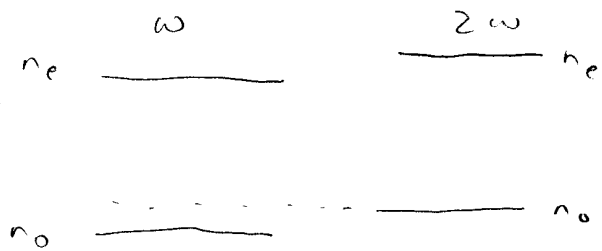
So if these components can't be resolved, have

$$P_{TOT} = P(2\omega_1) + P(2\omega_2) + P(\omega_1 + \omega_2)$$

$$= d \left\{ E(\omega_1)^2 + E(\omega_2)^2 + 2 E(\omega_1) E(\omega_2) \right\}$$

$$\boxed{P = d [E(\omega_1) + E(\omega_2)]^2} \quad \text{as desired}$$

3. By tilting crystal, can adjust  $n_e'$  between  $n_o$  and  $n_e$



So want to use  
 $n_o(2\omega) = n_e'(\omega)$

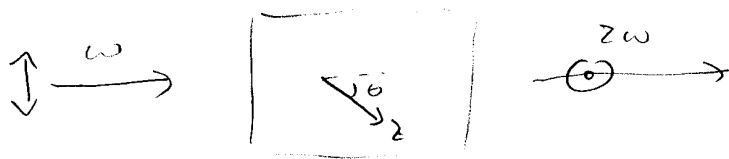
$$\text{So } \frac{\cos^2 \theta}{n_o(\omega)^2} + \frac{\sin^2 \theta}{n_e(\omega)^2} = \frac{1}{n_o(2\omega)^2} \quad \cos^2 \theta = 1 - \sin^2 \theta$$

$$\sin^2 \theta \left[ \frac{1}{n_e(\omega)^2} - \frac{1}{n_o(\omega)^2} \right] = \frac{1}{n_o(2\omega)^2} - \frac{1}{n_o(\omega)^2}$$

$$\sin^2 \theta = \frac{\frac{1}{n_o(2\omega)^2} - \frac{1}{n_o(\omega)^2}}{\frac{1}{n_e(\omega)^2} - \frac{1}{n_o(\omega)^2}} = \frac{\frac{1}{4.856^2} - \frac{1}{4.754^2}}{\frac{1}{6.243^2} - \frac{1}{4.754^2}} = 0.0618$$

$$\boxed{\theta = 14.4^\circ}$$

So we have extra-ordinary beam at  $\omega$ , and ordinary beam at  $2\omega$



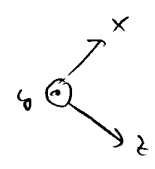
Need to  
 specify  $x, y$

Coupling constants are

$$\begin{aligned} d_{11} = d_{xxx} & : E_x(2\omega) \propto E_x(\omega)^2 \\ d_{12} = d_{xyy} & : E_x(2\omega) \propto E_y(\omega)^2 \\ d_{26} = d_{yxy} & : E_y(2\omega) \propto 2E_x(\omega)E_y(\omega) \end{aligned}$$

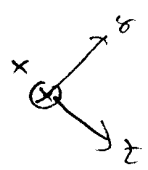
Check simple cases:

if crystal axes are



Have  $E_x(\omega) + E_y(2\omega)$   
doesn't work.

if axes are



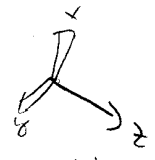
have  $E_y(\omega) = \cos\theta E(\omega)$  and  $E_x(2\omega) = E(2\omega)$

works with  $d_{12}$ :

$$d' = \frac{1}{2} d_{12} \cos^2\theta$$

$\uparrow$   
( $\omega_1 = \omega_2$ )

Just to be sure, check for  $x$  at angle  $\phi$   
out of page:



Take  $E(\omega) = \uparrow = E_x(\omega)$   
 $E(2\omega) = \odot = E_y(2\omega)$

Then

$$E_x(\omega) = \cos\phi \cos\theta E(\omega)$$
$$E_y(\omega) = -\sin\phi \cos\theta E(\omega)$$
$$P_y(2\omega) = \cos\phi P_y(2\omega) + \sin\phi P_x(2\omega)$$

$\omega_1 = \omega_2$  factor  
 $\downarrow$

Then

$$d' = \frac{1}{2} [d_{xxx} (\sin\phi \cdot \cos^2\phi \cos^2\theta) + d_{xyy} (-\sin\phi \sin^2\phi \cos^2\theta) + 2d_{yxx} (-\cos^2\phi \sin\phi \cos^2\theta)]$$

$$(d_{12} = -d_{11})$$
$$(d_{21} = -d_{11})$$

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$$d' = \frac{1}{2} d_{11} \sin \phi \cos^2 \theta [-\cos^2 \phi - \sin^2 \phi + 2 \cos^2 \phi]$$

$$= \frac{1}{2} d_{11} \sin \phi \cos^2 \theta (4 \cos^2 \phi - 1)$$

$$\sin \phi (4 \cos^2 \phi - 1) = 2 \sin \phi \cos^2 \phi + \sin \phi (2 \cos^2 \phi - 1)$$

$$= \sin 2\phi \cos \phi + \sin \phi \cos 2\phi$$

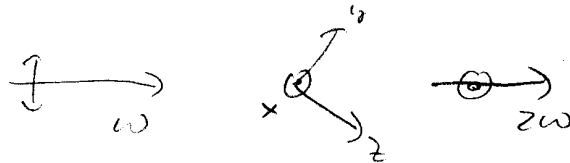
$$= \sin 3\phi$$

So  $d' = \frac{1}{2} d_{11} \cos^2 \theta \sin 3\phi$

Best at  $\phi = \pm 60^\circ, \pm 90^\circ$

Best case,  $|d'| = \frac{1}{2} d_{11} \cos^2 \theta = 2.67 \times 10^{-21} \frac{C}{V^2}$

Simplest is  $\phi = 90^\circ$ , as originally considered



4. From Dmitriev, Gurzadyan + Nikogosyan & Yarov

Symmetry class  $3m$ , uniaxial.  $d_{22} = -d_{21} = -d_{16}$ ,  $d_{24} = d_{15} \approx d_{31} = d_{32}$ ,  $d_{33}$

Transmission range 200 nm to 3.5  $\mu$ m

$$d_{22} = 2 \times 10^{-23} \frac{C}{V^2}$$

$$d_{31} = 1.4 \times 10^{-24} \frac{C}{V^2}$$

$$d_{33} = ?$$

$\lambda$	$n_o$	$n_e$
532 nm	1.6755	1.5549
1064 nm	1.6551	1.5425

5. Have  $\vec{E}(t) = \vec{E}(0) + \text{Re } \vec{E}(\omega) e^{i\omega t} = \vec{E}(0) + \frac{1}{2} \vec{E}(\omega) e^{i\omega t} + \frac{1}{2} \vec{E}^*(\omega) e^{-i\omega t}$  (7)

$$P_i(t) = \sum_{jk} 2d_{ijk} E_j(t) E_k(t)$$

$$= \sum_{jk} 2d_{ijk} \left( E_j(0) + \frac{1}{2} E_j(\omega) e^{i\omega t} + \frac{1}{2} E_j(\omega)^* e^{-i\omega t} \right) \times \left( E_k(0) + \frac{1}{2} E_k(\omega) e^{i\omega t} + \frac{1}{2} E_k(\omega)^* e^{-i\omega t} \right)$$

Component of  $P_i$  at  $\omega$  is

$$\frac{1}{2} [P_i(\omega) e^{i\omega t} + P_i^*(\omega) e^{-i\omega t}]$$

$$= \sum_{jk} 2d_{ijk} \left[ \frac{1}{2} E_j(0) E_k(\omega) e^{i\omega t} + \frac{1}{2} E_j(\omega) E_k(0) e^{i\omega t} + \frac{1}{2} E_j(0) E_k^*(\omega) e^{-i\omega t} + \frac{1}{2} E_j^*(\omega) E_k(0) e^{-i\omega t} \right]$$

So  $P_i(\omega) = \sum_{jk} 2d_{ijk} [E_j(0) E_k(\omega) + E_j(\omega) E_k(0)]$

$$= \sum_{jk} 4d_{ijk} E_j(\omega) E_k(0)$$

Since  $d_{ijk} = d_{ikj}$

So if we define  $\Delta\chi_{ij} \equiv \frac{1}{\epsilon_0} \sum_k 4d_{ijk} E_k(0)$ , have contribution

$$P_i(\omega) = \epsilon_0 \sum_j \Delta\chi_{ij} E_j(\omega)$$

So linear susceptibility is changed by  $\Delta\chi_{ij}$

Now  $r_{ijk}$  defined by

$$\Delta\chi_{ij} = \sum_k r_{ijk} E_k(0)$$

So we need to relate  $\Delta x_{ij}$  to  $\Delta y_{ij}$

$$\text{Have } \bar{y} = \epsilon_0 \bar{\Sigma}^{-1}$$

$$\bar{\Sigma} = \bar{I} + \bar{X}$$

$$\text{So } \bar{y} = \epsilon_0 (\bar{I} + \bar{X})^{-1}$$

In general, if  $B = A^{-1}$

and  $A$  is modified by  $\Delta A$ , how does  $B$  change?

$$\text{Have } AB = I$$

$$\text{So } (A + \Delta A)(B + \Delta B) = I$$

$$AB + (\Delta A)B + A(\Delta B) + (\Delta A)(\Delta B) = I$$

Assume  $(\Delta A)(\Delta B)$  is negligibly small

$$\text{Also, } AB = I, \text{ so}$$

$$(\Delta A)B + A(\Delta B) = 0$$

$$\Delta B = -A^{-1}(\Delta A)B$$

$$\Delta B = -B(\Delta A)B$$

$$\text{So here } \Delta y = -y \Delta X y$$

$$\text{or } \Delta y_{ij} = -\sum_{kl} y_{ik} \Delta x_{kl} y_{lj}$$

$$\text{So } \sum_m r_{ijm} E_m(0) = -\sum_{kl} y_{ik} \frac{1}{\epsilon_0} \sum_m 4d_{klm} E_m(0) y_{lj}$$

$$\sum_m \left[ r_{ijm} + \frac{4}{\epsilon_0} \sum_{kl} y_{ik} d_{klm} y_{lj} \right] E_m(0) = 0$$



(9)

Or

$$\Gamma_{ijm} = -\frac{4}{\epsilon_0} \sum_{kl} \gamma_{ik} d_{klm} \gamma_{lj}$$

But for unmodified crystal,  $\gamma$  is diagonal

$$\gamma_{ii} = \frac{\epsilon_0}{\epsilon_{ii}}$$

So need  $k=i$  and  $l=j$

$$\Gamma_{ijm} = -\frac{4}{\epsilon_0} \frac{\epsilon_0}{\epsilon_{ii}} d_{ijm} \frac{\epsilon_0}{\epsilon_{jj}}$$

Or

$$\Gamma_{ijk} = -4\epsilon_0 \frac{d_{ijk}}{\epsilon_{ii} \epsilon_{jj}}$$