

# 第四章 非线性光学效应

- 4. 1 光在非线性介质中的传播
- 4. 2 非线性光学相互作用下的波动方程
- 4. 3 二阶非线性光学过程
- 4. 4 三阶及高阶非线性光学过程

## 4.1 光在非线性介质中的传播

非线性光学效应 —— 物质对于外加电磁场的响应并不是外加电磁场振幅的线性函数的光学现象。

非线性：多光子过程

■ 非线性极化

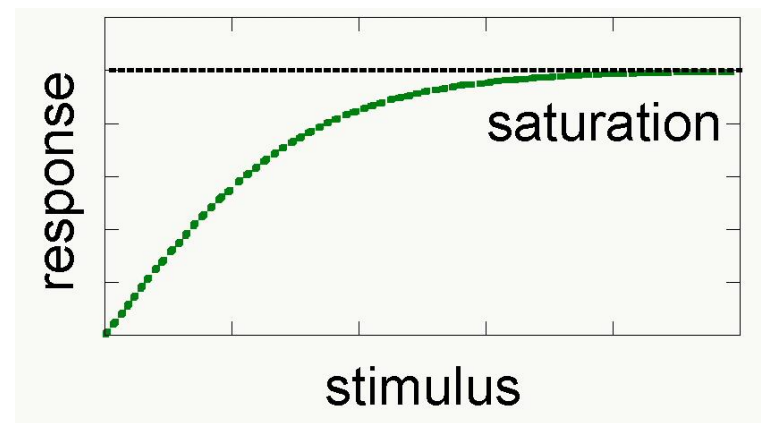
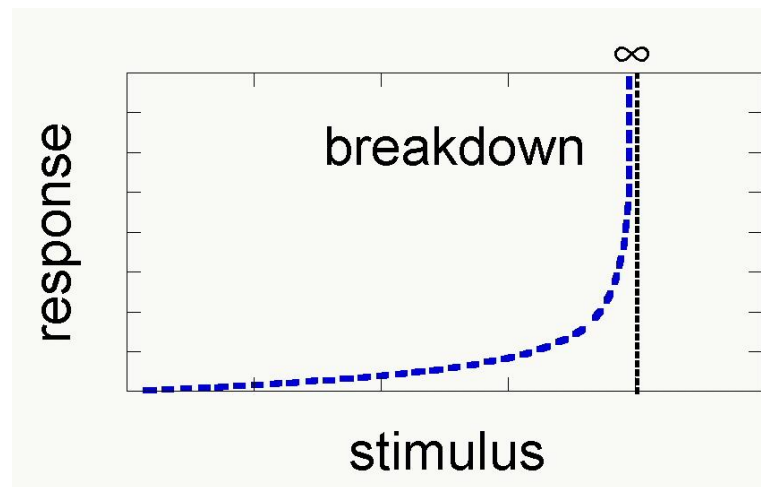
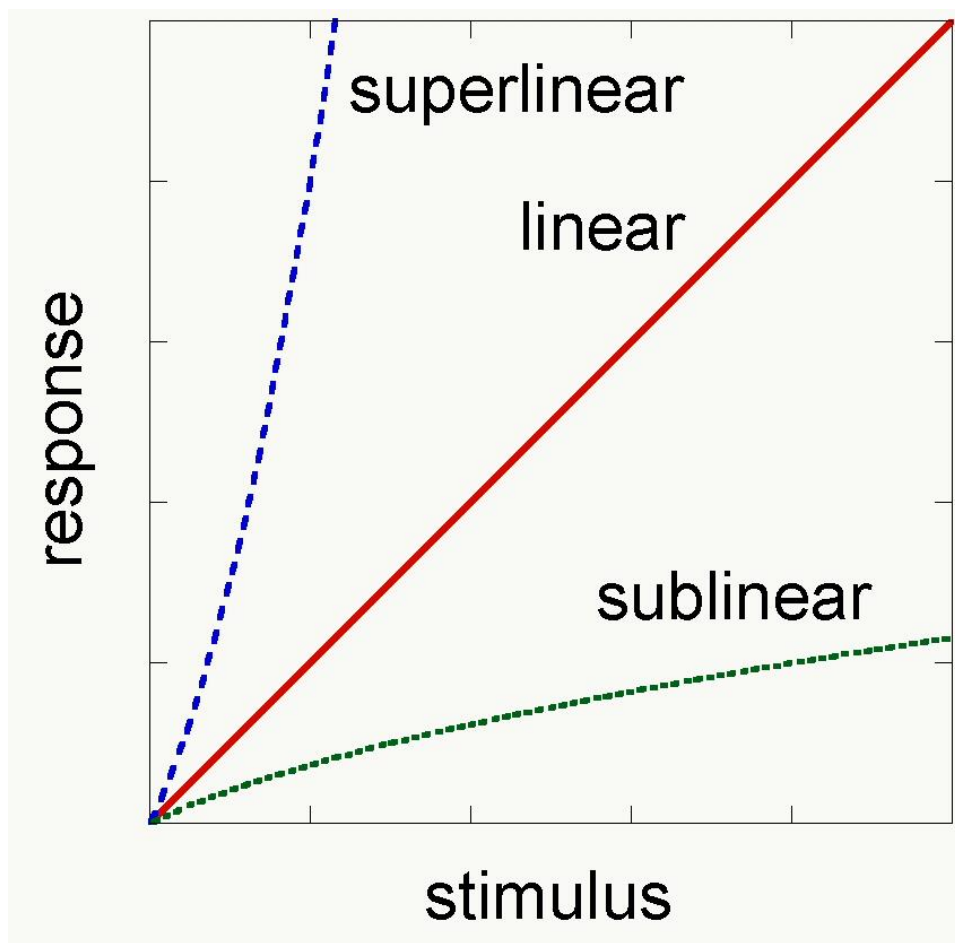
■ 二阶非线性效应：

二次谐波、和频、差频、参量过程；

■ 三阶非线性效应：

三次谐波、光克尔、自聚焦、四波混频、位相共轭

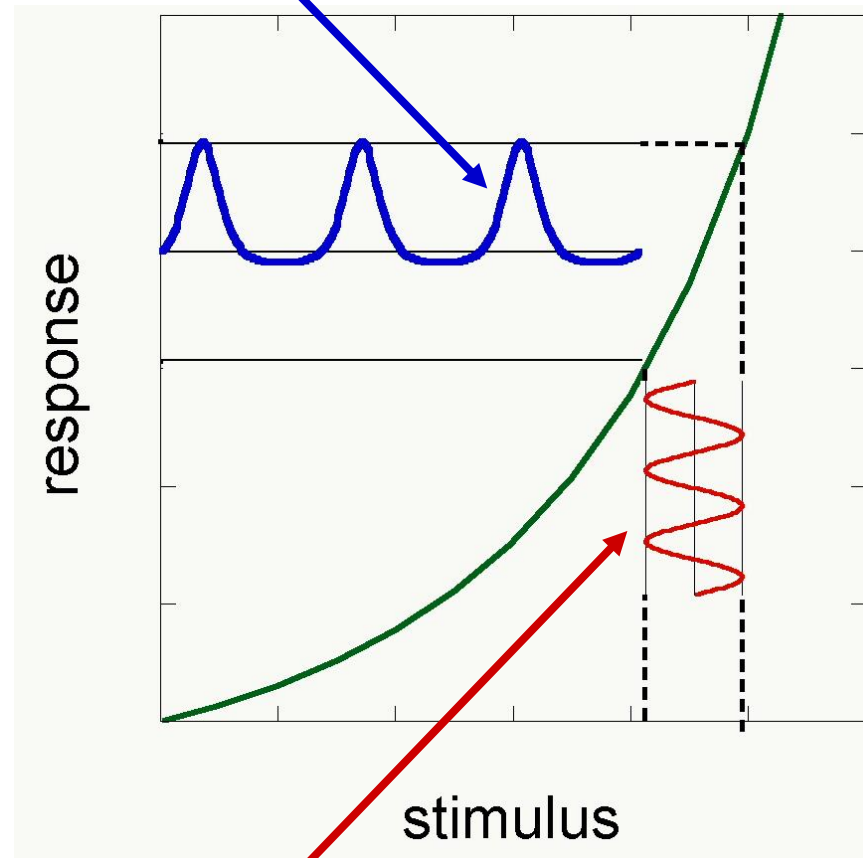
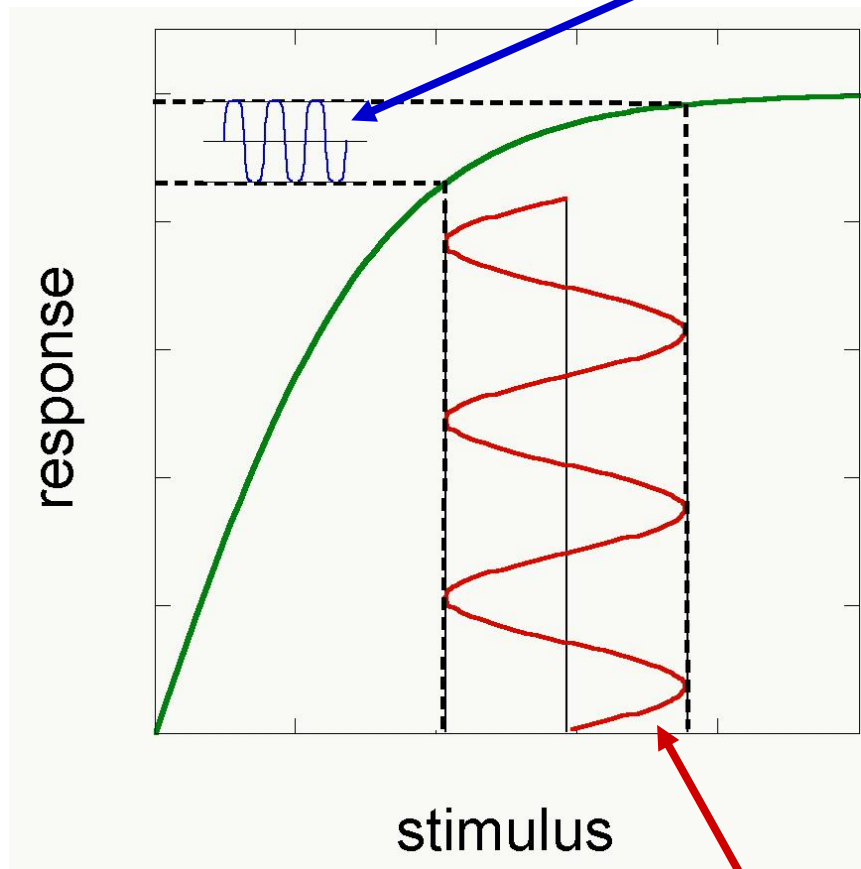
■ 高次谐波的产生



超线性和亚线性响应

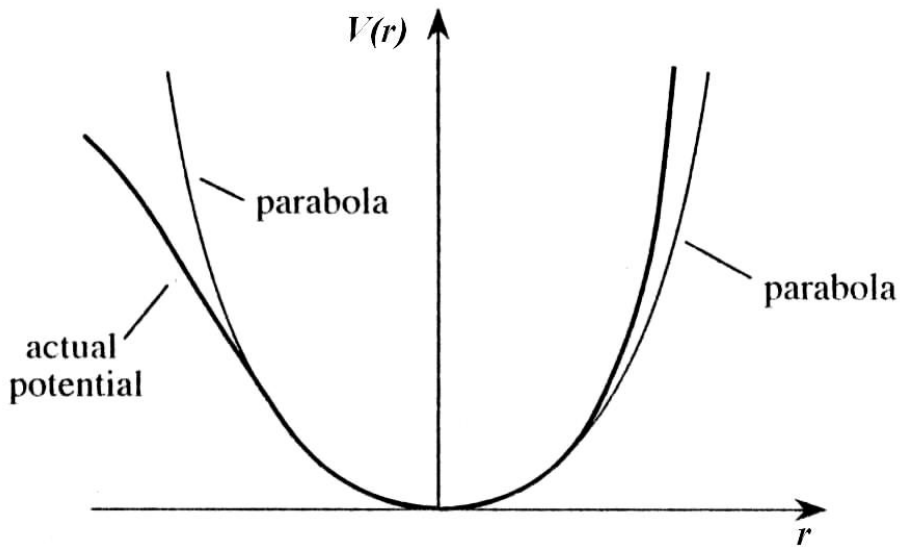
# 非线性的结果：产生新的频率

非正弦输出  
(混频)



正弦输入  
(单频)

# 产生非线性的原因：非谐振子



$$V(r) = \frac{1}{2}kr^2 + \frac{1}{3}\underline{D_1r^3} + \dots$$

非谐势能项

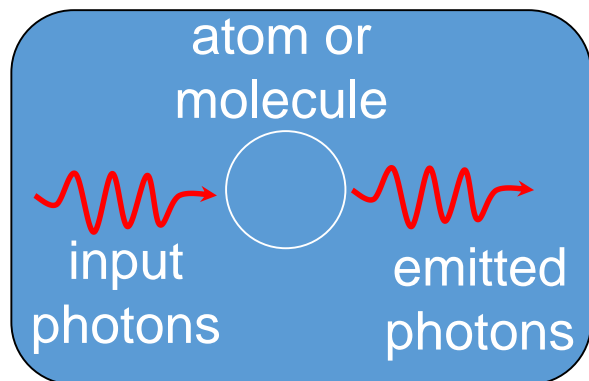
$$F(r) = -\frac{\partial V(r)}{\partial r} = -kr - \underline{D_1r^2} + \dots$$

非线性项

弱场 —— 谐振运动 —— 线性光学起主要作用

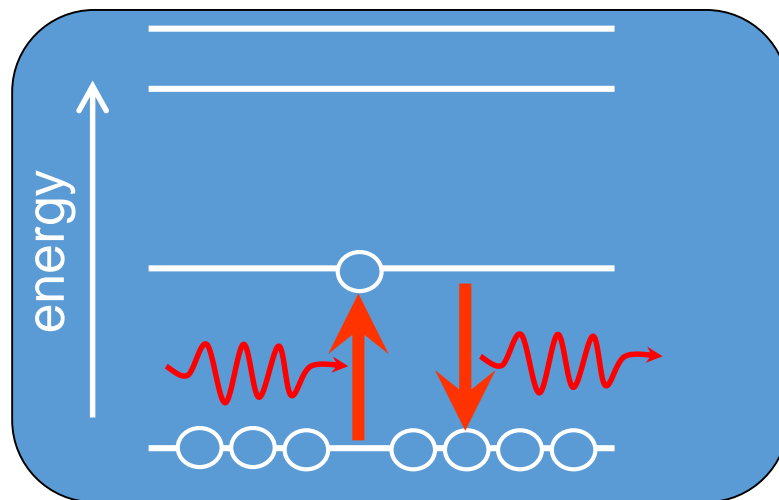
强场 —— 非谐运动 —— 产生新的频率

# 产生非线性的原因：高能级布居

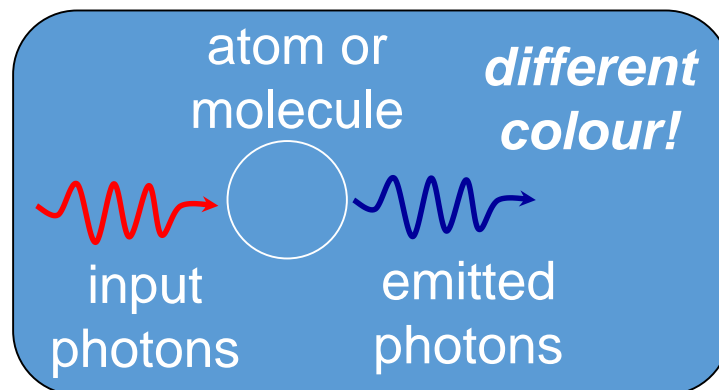
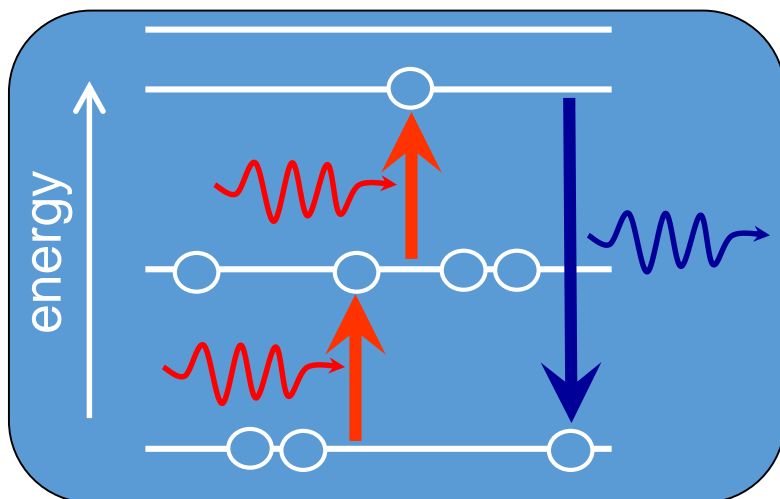


光引起分子中电子振动，  
产生辐射并与入射光相干

我们可以利用能级来  
表示这个过程



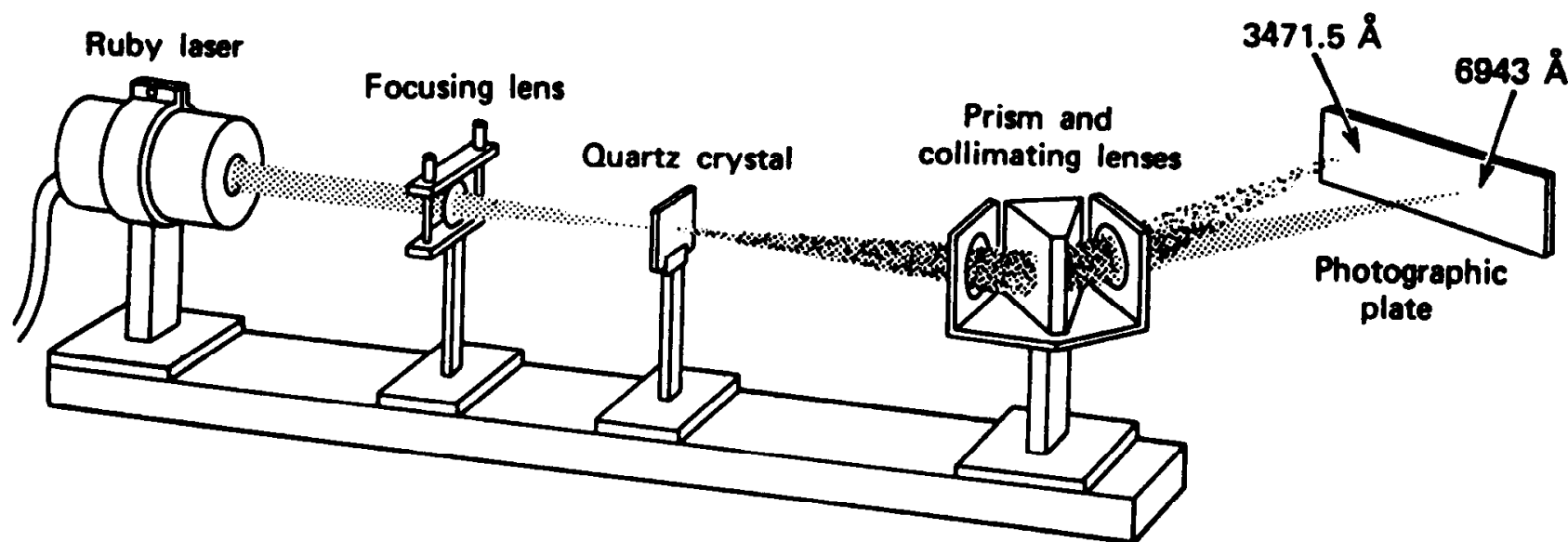
强入射光  $\rightarrow$  大的高能级布居数  $\rightarrow$  更高布居能级  $\rightarrow$  新的频率



非线性光学的发展——依赖于激光器的发展

# 首次二次谐波的产生

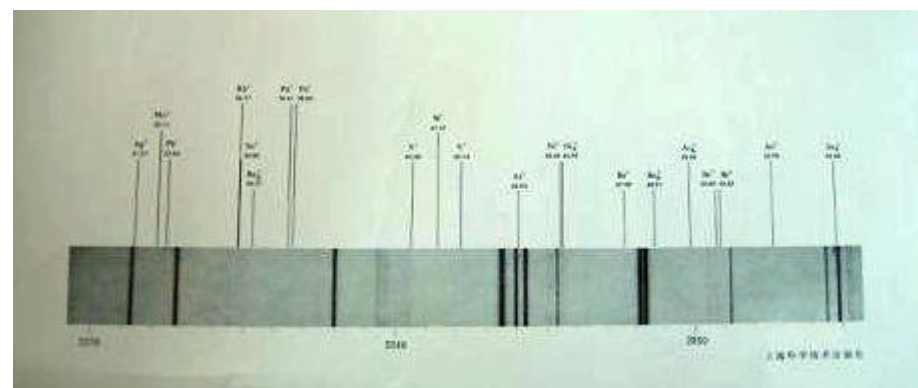
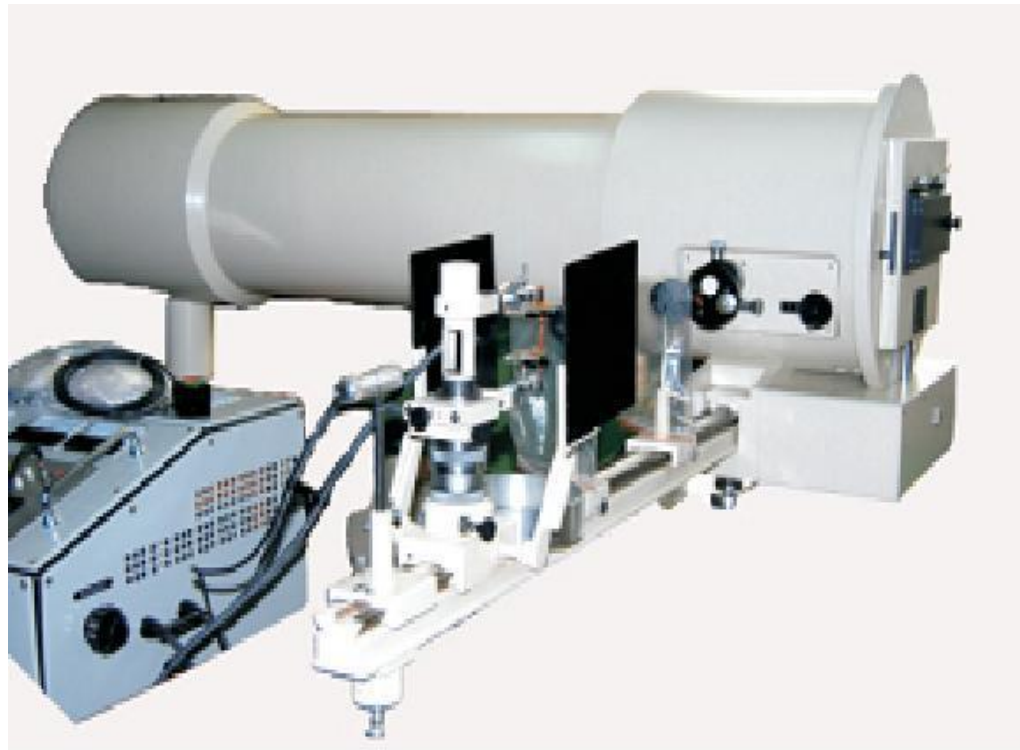
P.A. Franken et al, Phys. Rev. Lett. 7, 118 (1961)



**Figure 12.1.** Arrangement used in the first experimental demonstration of second-harmonic generation [1]. A ruby-laser beam at  $\lambda = 0.694 \mu\text{m}$  is focused on a quartz crystal, causing the generation of a (weak) beam at  $\frac{1}{2}\lambda = 0.347 \mu\text{m}$ . The two beams are then separated by a prism and detected on a photographic plate.

产生的二次谐波非常弱，因为没有实现相位匹配





# 当时的数据

实际发表的结果...

The second harmonic

Input beam

VOLUME 7, NUMBER 4

PHYSICAL REVIEW LETTERS

AUGUST 15, 1961

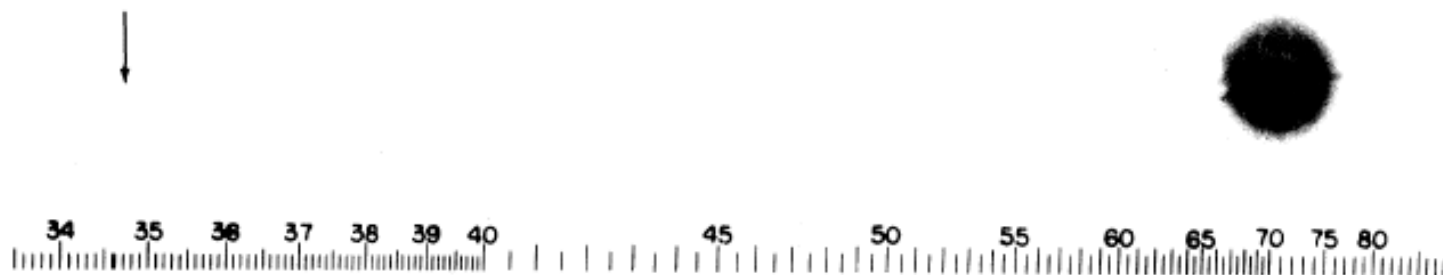


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 Å. The arrow at 3472 Å indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 Å is very large due to halation.

注意：弱的二次谐波的点丢失了！

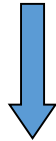
## 4.1.1 非线性光学过程的描述

非线性光学过程包含两个部分：

- 强光导致的介质非线性响应：介质性质方程描述
- 介质对光的非线性调制：波动方程描述

所有介质都可以表现出非线性效应，材料不同效应的大小也不同。

极化强度:  $P = \varepsilon_0 \chi E$



非线性极化

$$P = \varepsilon_0 [\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots]$$

结果:  $E(t) \propto E_0 \exp(-i\omega t) + E_0^* \exp(i\omega t)$

$$E(t)^2 \propto E_0^2 \exp(-2i\omega t) + 2|E_0|^2 + E_0^{*2} \exp(2i\omega t)$$



$2\omega = 2\text{nd harmonic!}$

$$\mathbf{P} = \varepsilon_0 \boldsymbol{\chi}^{(1)} \cdot \mathbf{E} + \varepsilon_0 \boldsymbol{\chi}^{(2)} : \mathbf{E}\mathbf{E} + \varepsilon_0 \boldsymbol{\chi}^{(3)} : \mathbf{E}\mathbf{E}\mathbf{E} + \dots$$

$$= \mathbf{P}^{(1)} + \mathbf{P}^{(2)} + \mathbf{P}^{(3)} + \dots$$

$$= \mathbf{P}^{(1)} + \mathbf{P}_{NL}$$

$$\mathbf{P}_{NL} = \mathbf{P}^{(2)} + \mathbf{P}^{(3)} + \dots$$

非线性极化强度
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$$\mathbf{P}^{(1)} = \varepsilon_0 \boldsymbol{\chi}^{(1)} \cdot \mathbf{E} \quad (\text{线性效应})$$

$$\mathbf{P}^{(2)} = \varepsilon_0 \boldsymbol{\chi}^{(2)} : \mathbf{E}\mathbf{E} \quad (\text{二阶非线性效应})$$

$$\mathbf{P}^{(3)} = \varepsilon_0 \boldsymbol{\chi}^{(3)} : \mathbf{E}\mathbf{E}\mathbf{E} \quad (\text{三阶非线性效应})$$

$\boldsymbol{\chi}^{(1)}$ ——线性极化率，二阶张量，9个元素

$\boldsymbol{\chi}^{(2)}$ ——二阶非线性极化率，三阶张量，27个元素

$\boldsymbol{\chi}^{(3)}$ ——三阶非线性极化率，四阶张量，81个元素

$$\left| \frac{P^{(n+1)}}{P^{(n)}} \right| \sim \left| \frac{E}{E_{atom}} \right|$$

$$E_{atom} \sim 3 \times 10^{10} \text{V/m}$$

观察非线性效应需要： 1kV/cm  $\sim$  2.5 kW/cm<sup>2</sup>

# 单位制

MKS/SI单位制:

$$P^{(n)} = \varepsilon_0 \chi^{(n)} E^n$$

$$\varepsilon_0 = 8.85 \times 10^{-12} \text{ F / m}$$

$$[P^{(n)}] = \text{Cm}^{-2}$$

$$[\chi^{(n)}] = (\text{mV}^{-1})^{n-1}$$

$$[E] = \text{Vm}^{-1}$$

cgs/esu单位制:

$$P^{(n)} = \chi^{(n)} E^n$$

$$[P^{(n)}] = \text{cm}^{-\frac{1}{2}} \text{g}^{\frac{1}{2}} \text{s}^{-1}$$

$$[\chi^{(n)}] = (\text{cm}^{\frac{1}{2}} \text{g}^{-\frac{1}{2}} \text{s})^{n-1}$$

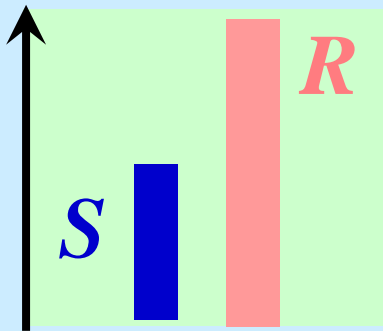
$$[E] = \text{cm}^{-\frac{1}{2}} \text{g}^{\frac{1}{2}} \text{s}^{-1}$$

换算关系:  $\chi^{(n)}(\text{SI}) / \chi^{(n)}(\text{esu}) = 4\pi / (10^{-4} c)^{n-1}$

 真空光速

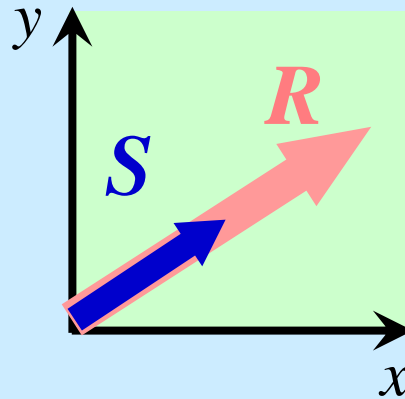
# 矢量的激励—响应函数

## Scalars



$$R = \zeta S$$
$$\zeta = \text{constant}$$

## Parallel vectors

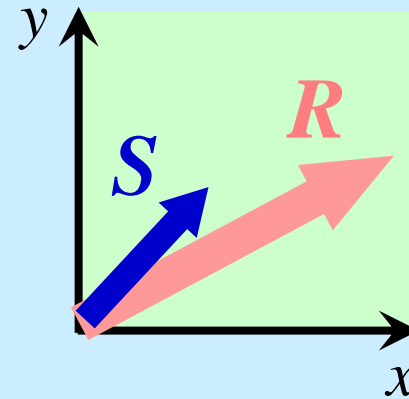


$$\underline{R} = \zeta \underline{S}$$
$$\zeta = \text{constant}$$

$$\begin{pmatrix} R_x \\ R_y \\ R_z \end{pmatrix} = \zeta \begin{pmatrix} S_x \\ S_y \\ S_z \end{pmatrix}$$

isotropic

## Nonparallel vectors



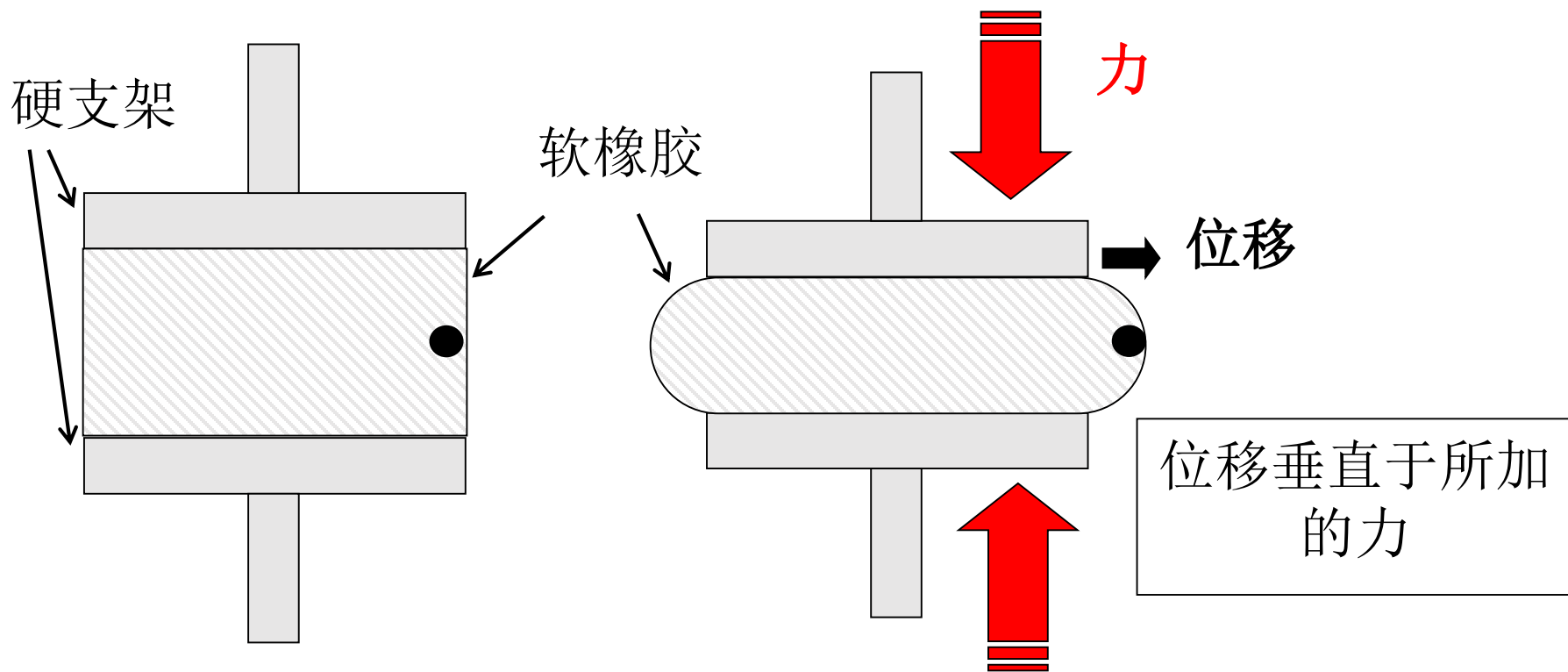
$$\underline{R} = \zeta \underline{S}$$
$$\zeta = \text{tensor}$$

$$\begin{pmatrix} R_x \\ R_y \\ R_z \end{pmatrix} = \begin{pmatrix} \zeta_{xx} & \zeta_{xy} & \zeta_{xz} \\ \zeta_{yx} & \zeta_{yy} & \zeta_{yz} \\ \zeta_{zx} & \zeta_{zy} & \zeta_{zz} \end{pmatrix} \begin{pmatrix} S_x \\ S_y \\ S_z \end{pmatrix}$$

anisotropic



# 例：挤压橡胶

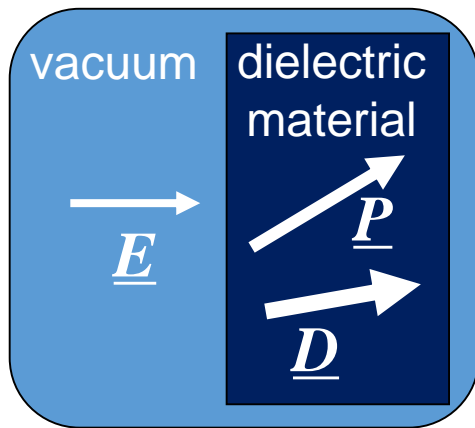


弹性系数是张量，对于各向异性材料有36个分量

# 各向异性介质中的光

对于各向异性介质 (大多数晶体; 电场中的各向同性介质):

- **D, P和E不平行**
- $\chi$  和  $\epsilon_r$  为二秩张量
- 光速和折射率是各向异性的:  
随传播方向和偏振不同而不同



$$\mathbf{P} = \overline{\overline{\chi}} \epsilon_0 \mathbf{E}$$

$$\overline{\overline{\chi}} = \text{susceptibility tensor} = \begin{pmatrix} \chi_{xx} & \chi_{xy} & \chi_{xz} \\ \chi_{yx} & \chi_{yy} & \chi_{yz} \\ \chi_{zx} & \chi_{zy} & \chi_{zz} \end{pmatrix}$$

$$\mathbf{D} = \overline{\overline{\epsilon_r}} \epsilon_0 \mathbf{E}$$

$$\overline{\overline{\epsilon_r}} = \text{dielectric tensor} = \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\ \epsilon_{yx} & \epsilon_{yy} & \epsilon_{yz} \\ \epsilon_{zx} & \epsilon_{zy} & \epsilon_{zz} \end{pmatrix}$$

# 各向异性介电常数

• **D**, **P**和**E**不平行

$$\mathbf{D} = \varepsilon_0 \bar{\bar{\varepsilon}}_r \mathbf{E}$$

下列下标是等价的：

$$\begin{pmatrix} D_x \\ D_y \\ D_z \end{pmatrix} = \varepsilon_0 \begin{pmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ \varepsilon_{yx} & \varepsilon_{yy} & \varepsilon_{yz} \\ \varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$

$$\begin{pmatrix} D_1 \\ D_2 \\ D_3 \end{pmatrix} = \varepsilon_0 \begin{pmatrix} \varepsilon_{11} & \varepsilon_{12} & \varepsilon_{13} \\ \varepsilon_{21} & \varepsilon_{22} & \varepsilon_{23} \\ \varepsilon_{31} & \varepsilon_{32} & \varepsilon_{33} \end{pmatrix} \begin{pmatrix} E_1 \\ E_2 \\ E_3 \end{pmatrix}$$

$$\begin{cases} D_x = \varepsilon_0 \varepsilon_{xx} E_x + \varepsilon_0 \varepsilon_{xy} E_y + \varepsilon_0 \varepsilon_{xz} E_z \\ D_y = \varepsilon_0 \varepsilon_{yx} E_x + \varepsilon_0 \varepsilon_{yy} E_y + \varepsilon_0 \varepsilon_{yz} E_z \\ D_z = \varepsilon_0 \varepsilon_{zx} E_x + \varepsilon_0 \varepsilon_{zy} E_y + \varepsilon_0 \varepsilon_{zz} E_z \end{cases}$$

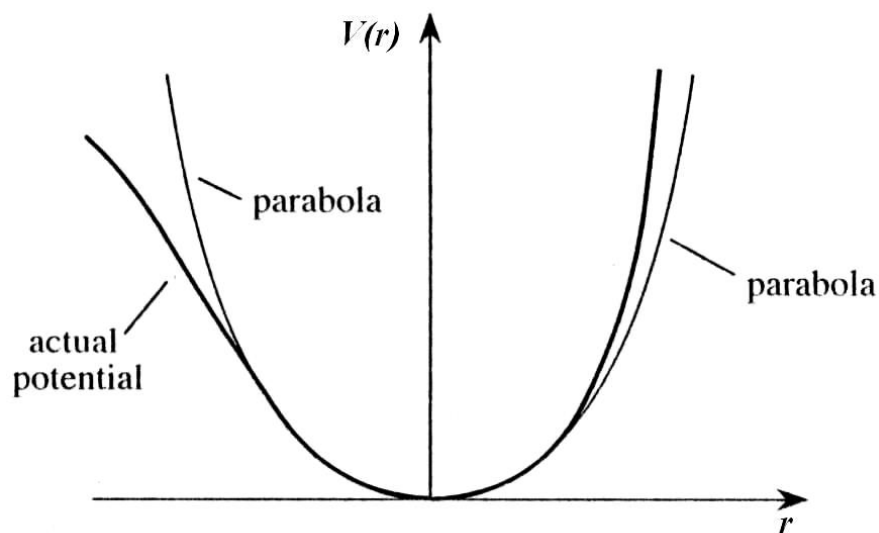
$$\begin{cases} D_1 = \varepsilon_0 \varepsilon_{11} E_1 + \varepsilon_0 \varepsilon_{12} E_2 + \varepsilon_0 \varepsilon_{13} E_3 \\ D_2 = \varepsilon_0 \varepsilon_{21} E_1 + \varepsilon_0 \varepsilon_{22} E_2 + \varepsilon_0 \varepsilon_{23} E_3 \\ D_3 = \varepsilon_0 \varepsilon_{31} E_1 + \varepsilon_0 \varepsilon_{32} E_2 + \varepsilon_0 \varepsilon_{33} E_3 \end{cases}$$

$$D_i = \sum_{j=1}^3 \varepsilon_0 \varepsilon_{ij} E_j$$

## 二阶非线性极化率张量元

$$\begin{bmatrix} \chi_{xxx} & \chi_{xyy} & \chi_{xzz} & \chi_{xyz} & \chi_{xzy} & \chi_{xzx} & \chi_{xxz} & \chi_{xxy} & \chi_{xyx} \\ \chi_{yxx} & \chi_{yyy} & \chi_{yzz} & \chi_{yyz} & \chi_{yzy} & \chi_{yzx} & \chi_{yxz} & \chi_{yxy} & \chi_{yyx} \\ \chi_{zxx} & \chi_{zyy} & \chi_{zzz} & \chi_{zyz} & \chi_{zzy} & \chi_{zzx} & \chi_{zxz} & \chi_{zxy} & \chi_{zyx} \end{bmatrix}$$

## 4.1.2 非线性极化率的求解



$$V(r) = \frac{1}{2}kr^2 + \frac{1}{3}D_1r^3$$

$$F(r) = -\frac{\partial V(r)}{\partial r} = -kr - D_1r^2$$


电子的一维势场

$-\omega \text{项: } E^*(\omega) = E(-\omega)$

光电场:  $E = E(\omega)e^{-i\omega t} + E^*(\omega)e^{i\omega t}$

电子运动方程:  $\ddot{r} + \gamma\dot{r} + \omega_0^2 r + Dr^2 = -\frac{e}{m}E \quad D = D_1/m$

$$P = P^{(1)} + P^{(2)} + P^{(3)} + \dots \quad \text{考虑微扰的情况: } P^{(1)} \gg P^{(2)} \gg P^{(3)}$$

$$r = r_1 + r_2 + r_3 + \dots \quad r_1 \sim E \quad r_2 \sim EE \quad r_3 \sim EEE$$


$$\ddot{r} + \gamma \dot{r} + \omega_0^2 r + Dr^2 = -\frac{e}{m} E$$

$$\begin{aligned} & \frac{d^2 r_1}{dt^2} + \gamma \frac{dr_1}{dt} + \omega_0^2 r_1 + Dr_1^2 + 2Dr_1 r_2 + Dr_2^2 \\ & + \frac{d^2 r_2}{dt^2} + \gamma \frac{dr_2}{dt} + \omega_0^2 r_2 + \dots = -\frac{e}{m} E \end{aligned}$$

$$\frac{d^2 r_1}{dt^2} + \gamma \frac{dr_1}{dt} + \omega_0^2 r_1 = -\frac{e}{m} E$$

$$\frac{d^2 r_2}{dt^2} + \gamma \frac{dr_2}{dt} + \omega_0^2 r_2 = -Dr_1^2$$

洛伦兹  
模型:

$$r_1(t) = -\frac{e}{m} \frac{E}{(\omega_0^2 - \omega^2) - i\gamma\omega}$$

$$F(\omega) = \frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega}$$

$$r_1(t) = -\frac{e}{m} E(\omega) F(\omega) e^{-i\omega t} + c.c.$$

$$r_2 = -\frac{De^2}{m^2} E^2(\omega) F(2\omega) F(\omega) F(\omega) e^{-i2\omega t} - \frac{De^2}{m^2} E(\omega) E^*(\omega) F(0) F(\omega) F(-\omega) + c.c.$$

$$P = -Ner = -Ne(r_1 + r_2 + \dots) = P^{(1)} + P^{(2)} + \dots$$

$$\left\{ \begin{aligned} P^{(1)}(t) &= \frac{Ne^2}{m} E(\omega) F(\omega) e^{-i\omega t} + c.c. \\ P^{(2)} &= \frac{DNe^3}{m^2} E^2(\omega) F(2\omega) F(\omega) F(\omega) e^{-i2\omega t} + \frac{DNe^3}{m^2} E(\omega) E^*(\omega) F(0) F(\omega) F(-\omega) + c.c. \end{aligned} \right.$$

新频率

$$\left\{ \begin{aligned} P^{(1)}(t) &= \varepsilon_0 \chi^{(1)}(-\omega, \omega) E(\omega) e^{-i\omega t} + c.c. \\ P^{(2)}(t) &= \varepsilon_0 \chi^{(2)}(-2\omega, \omega, \omega) E^2(\omega) e^{-i2\omega t} + \varepsilon_0 \chi^{(2)}(0, \omega, -\omega) E(\omega) E(-\omega) + c.c. \end{aligned} \right.$$

$$\chi^{(1)}(-\omega, \omega) = \frac{Ne^2}{\varepsilon_0 m} F(\omega)$$

$$\chi^{(2)}(-2\omega, \omega, \omega) = \frac{DNe^3}{\varepsilon_0 m^2} F(2\omega)F(\omega)F(\omega)$$

$$\chi^{(2)}(0, \omega, -\omega) = \frac{DNe^3}{\varepsilon_0 m^2} F(0)F(\omega)F(-\omega)$$

二阶极化率的一般形式：

$$\chi^{(2)}(-\omega_1 - \omega_2, \omega_1, \omega_2) = \frac{DNe^3}{\varepsilon_0 m^2} F(\omega_1 + \omega_2)F(\omega_1)F(\omega_2)$$

极化强度可以写成分量表表达式：

$$P_i^{(2)}(t) = \varepsilon_0 \sum_{\substack{m,n \\ j,k}} \chi_{ijk}^{(2)}(-\omega_m - \omega_n, \omega_m, \omega_n) E_j(\omega_m) E_k(\omega_n) e^{-i(\omega_m + \omega_n)t}$$

其中  $i, j, k = x, y, z$



倍频系数与二阶非线性极化率的关系：

$$d_{ijk}(-2\omega; \omega, \omega) = \frac{1}{2} \chi_{ijk}^{(2)}(-2\omega; \omega, \omega)$$

$(jk)$	11	22	33	23 32	13 31	12 21
$(\mu)$	1	2	3	4	5	6

$$d_{i\mu}(-2\omega; \omega, \omega) = \frac{1}{2} \chi_{ijk}^{(2)}(-2\omega; \omega, \omega)$$

晶 体	对称 类型	透明波段 / $\mu\text{m}$	折射率(20℃)			非线性系数 /(pm/V)
			波长 / $\mu\text{m}$	$n_o$	$n_e$	
$\text{LiNbO}_3$	$3m$	0.4~5	0.53	2.33	2.23	$d_{15}=5.45(1.06\mu\text{m})$ $d_{22}=2.76(1.06\mu\text{m})$
			0.694	2.28	2.19	
			1.06	2.23	2.16	
$\beta\text{-BaB}_2\text{O}_4$	3	0.19~3	1.064	1.66	1.54	$d_{11}=1.78(1.079\mu\text{m})$ $d_{22}=0.13(1.079\mu\text{m})$ $d_{31}=0.13(1.079\mu\text{m})$
			0.532	1.67	1.55	
		0.19~3	0.3547	1.70	1.58	
			0.2660	1.78	1.62	

### 4.1.3 非线性极化率的对称性

#### (1) 真实性条件

电场是实际物理量:  $\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_0(\mathbf{r}) \cos(\omega t + \varphi)$

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}(\omega)e^{-i\omega t} + \mathbf{E}^*(\omega)e^{i\omega t}$$

$\mathbf{E}(\omega)$ 为频域复振幅:  $\mathbf{E}(\omega) = \frac{1}{2} \mathbf{E}_0(\mathbf{r}) e^{-i\varphi(\mathbf{r})}$

$$\mathbf{E}^*(\omega) = \mathbf{E}(-\omega)$$

同理:  $\mathbf{P}^*(\omega) = \mathbf{P}(-\omega)$

频域:  $\mathbf{P}^{(1)}(\omega) = \varepsilon_0 \chi^{(1)}(\omega) \cdot \mathbf{E}(\omega)$

$$\begin{aligned} [\mathbf{P}^{(1)}(\omega)]^* &= \mathbf{P}^{(1)}(-\omega) = \varepsilon_0 [\chi^{(1)}(\omega)]^* \mathbf{E}^*(\omega) \\ &= \varepsilon_0 [\chi^{(1)}(\omega)]^* \mathbf{E}(-\omega) \end{aligned}$$

$$[\chi^{(1)}(\omega)]^* = \chi^{(1)}(-\omega) \quad \longrightarrow \quad \varepsilon^*(\omega) = \varepsilon(-\omega)$$

$$[\chi^{(2)}(\omega_1, \omega_2)]^* = \chi^{(2)}(-\omega_1, -\omega_2)$$

$$[\chi^{(3)}(\omega_1, \omega_2, \omega_3)]^* = \chi^{(3)}(-\omega_1, -\omega_2, -\omega_3)$$

## (2)本征置换对称性

光场等价：数学上的作用顺序可变

$$\varepsilon_0 \chi_{ijk}^{(2)}(-\omega_1 - \omega_2, \omega_1, \omega_2) E_j(\omega_1) E_k(\omega_2) e^{-i(\omega_1 + \omega_2)t}$$

$$\varepsilon_0 \chi_{ikj}^{(2)}(-\omega_1 - \omega_2, \omega_2, \omega_1) E_k(\omega_2) E_j(\omega_1) e^{-i(\omega_1 + \omega_2)t}$$

$$\chi_{ijk}^{(2)}(-\omega_1 - \omega_2, \omega_1, \omega_2) = \chi_{ikj}^{(2)}(-\omega_1 - \omega_2, \omega_2, \omega_1)$$

### (3)全对称性

$$F(\omega) = \frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega}$$

$$F(\omega) = \frac{\omega_0^2 - \omega^2}{(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2} + i \frac{\gamma\omega}{(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2}$$

所有 $\omega$ 在透明区域，忽略虚部： $F(\omega) = F(-\omega)$

$$\chi^{(2)}(\omega_1, -\omega_1 - \omega_2, \omega_2) \propto F(-\omega_1)F(-\omega_1 - \omega_2)F(\omega_2)$$

$$\chi^{(2)}(-\omega_1 - \omega_2, \omega_1, \omega_2) \propto F(\omega_1 + \omega_2)F(\omega_1)F(\omega_2)$$

$$\chi^{(2)}(\omega_1, -\omega_1 - \omega_2, \omega_2) = \chi^{(2)}(\omega_2, \omega_1, -\omega_1 - \omega_2) = \chi^{(2)}(-\omega_1 - \omega_2, \omega_1, \omega_2)$$

#### (4) Kleinman对称性

$$\omega \ll \omega_0: \quad F(\omega) = \frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega} \approx \frac{1}{\omega_0^2}$$

$$\chi^{(2)}(-\omega_1 - \omega_2, \omega_1, \omega_2) = \chi^{(2)}(-\omega_1' - \omega_2', \omega_1', \omega_2')$$

## (5) 空间对称性

具有反演中心的晶体没有偶数阶极化率张量

$$\mathbf{P}^{(2)}(t) = \sum_{m,n} \varepsilon_0 \chi^{(2)} : \mathbf{E}(\omega_m) \mathbf{E}(\omega_n) e^{-i(\omega_m + \omega_n)t}$$

$x \rightarrow -x, y \rightarrow -y, z \rightarrow -z$  的坐标变换下,  $\mathbf{E}$  和  $\mathbf{P}$  都改变了方向

$$-\mathbf{P}^{(2)}(t) = \mathbf{P}^{(2)}(t) \longrightarrow \mathbf{P}^{(2)}(t) = 0$$

$$\mathbf{E}(\omega_m), \mathbf{E}(\omega_n) \text{ 不为零} \longrightarrow \chi^{(2)} = 0$$



## 1. 三斜晶系

晶类  $1(C_1)$

$$\begin{bmatrix} XXX & XYY & XZZ & XYZ & XZY & XZX & XXZ & XXY & XYX \\ YXX & YYY & YZZ & YYZ & YZY & YZX & YXZ & YXY & YYX \\ ZXX & ZYY & ZZZ & ZYZ & ZZY & ZZX & ZXZ & ZXY & ZYX \end{bmatrix}$$

## 2. 单斜晶系

晶类  $2(C_2)$

$$\begin{bmatrix} 0 & 0 & 0 & XYZ & XZY & 0 & 0 & XXY & XYX \\ YXX & YYY & YZZ & 0 & 0 & YZX & YXZ & 0 & 0 \\ 0 & 0 & 0 & ZYZ & ZZY & 0 & 0 & ZXY & ZYX \end{bmatrix}$$

晶类  $m(C_s)$

$$\begin{bmatrix} XXX & XYY & XZZ & 0 & 0 & XZX & XXZ & 0 & 0 \\ 0 & 0 & 0 & YYZ & YZY & 0 & 0 & YXY & YYX \\ ZXX & ZYY & ZZZ & 0 & 0 & ZZX & ZXZ & 0 & 0 \end{bmatrix}$$

## 4.1.4 非线性波动方程

$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}^{(1)} + \mathbf{P}_{NL}$  代入波动方程

$$\nabla^2 \mathbf{E} - \sigma \mu \frac{\partial \mathbf{E}}{\partial t} - \varepsilon \mu \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2}$$

根据 $\mathbf{P}_{NL}$ 的模式（偏振、频率、传播方向），分为两类：

1. 非线性频率变换：极化强度的模式与入射的多个电磁场的模式都不一样。包括谐波产生、和频、差频、参量放大与振荡、相干喇曼散射，光整流等。

2. 非线性吸收和场感生折射率变化：极化强度的模式与某一入射电磁场的模式一样。包括双光子吸收和多光子吸收、电光效应、自作用过程、场致双折射等。

## 4.2 二阶非线性光学过程

### 4.2.1 非线性耦合波方程

### 4.2.2 光的二次谐波

### 4.2.3 其它二阶非线性光学现象

## 4.2.1 非线性耦合波方程

$$\nabla^2 \mathbf{E} - \sigma \mu \frac{\partial \mathbf{E}}{\partial t} - \varepsilon \mu \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2}$$

$$\mathbf{E}(\mathbf{r}, t) = \sum_n \mathbf{E}_n(\mathbf{r}, t) \quad \mathbf{P}^{NL}(\mathbf{r}, t) = \sum_n \mathbf{P}_n^{NL}(\mathbf{r}, t)$$

$$\mathbf{E}_n(z, t) = \mathbf{E}_n(z) e^{ik_n z - i\omega_n t} + c.c. \quad \mathbf{P}_n^{NL}(z, t) = \mathbf{P}_n^{NL}(z) e^{ik'_n z - i\omega_n t} + c.c.$$

↑  
假设稳态：振幅不随时间变化  
↑

$k_n$ : 光的波数;  $k'_n$ : 极化的波数; 两者不一定相等

$$\nabla^2 \mathbf{E} - \varepsilon \mu \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2}$$

$$\mathbf{E}_n(z, t) = \mathbf{E}_n(z) e^{ik_n z - i\omega_n t} \quad \mathbf{P}_n^{NL}(z, t) = \mathbf{P}_n^{NL}(z) e^{ik_n' z - i\omega_n t}$$

$$\frac{\partial}{\partial z} \left[ \frac{\partial \mathbf{E}_n}{\partial z} e^{i(k_n z - \omega_n t)} + ik_n \mathbf{E}_n e^{i(k_n z - \omega_n t)} \right] + \mu_0 \varepsilon \omega_n^2 \mathbf{E}_n e^{i(k_n z - \omega_n t)} = -\mu_0 \omega_n^2 \mathbf{P}_n^{NL} e^{i(k_n' z - \omega_n t)}$$

$$\frac{\partial^2 \mathbf{E}_n}{\partial z^2} + ik_n \frac{\partial \mathbf{E}_n}{\partial z} + ik_n \frac{\partial \mathbf{E}_n}{\partial z} - k_n^2 \mathbf{E}_n + \mu_0 \varepsilon \omega_n^2 \mathbf{E}_n = -\mu_0 \omega_n^2 \mathbf{P}_n^{NL} e^{i\Delta k z}$$

$$\Delta k = k_n' - k_n \quad k_n^2 = \frac{n^2 \omega_n^2}{c^2} = \varepsilon_r \mu_0 \varepsilon_0 \omega_n^2 = \varepsilon \mu_0 \omega_n^2$$

$$\frac{\partial^2 \mathbf{E}_n}{\partial z^2} + 2ik_n \frac{\partial \mathbf{E}_n}{\partial z} = -\mu_0 \omega_n^2 \mathbf{P}_n^{NL} e^{i\Delta k z}$$

$$\frac{\partial^2 \mathbf{E}_n}{\partial z^2} + 2ik_n \frac{\partial \mathbf{E}_n}{\partial z} = -\mu_0 \omega_n^2 \mathbf{P}_n^{NL} e^{i\Delta k z}$$

采用空间的慢变振幅近似：

$$\left| \frac{\partial^2 \mathbf{E}_n}{\partial z^2} \right| \ll \left| \mathbf{k}_n \cdot \frac{\partial \mathbf{E}_n}{\partial z} \right|$$

稳态条件下非线性耦合波方程：

$$\frac{\partial \mathbf{E}_n}{\partial z} = \frac{i\omega_n}{2\varepsilon_0 c n} \mathbf{P}_n^{NL} e^{i\Delta k z}$$

$$\nabla^2 \mathbf{E} - \varepsilon\mu \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2}$$

$$\mathbf{E}_n(z, t) = \mathbf{E}_n(z, t) e^{ik_n z - i\omega_n t} \quad \mathbf{P}_n^{NL}(z, t) = \mathbf{P}_n^{NL}(z, t) e^{ik'_n z - i\omega_n t}$$

光电场振幅随时间变化时：

$$\begin{aligned} -\varepsilon\mu_0 \frac{\partial^2 \mathbf{E}_n(z, t)}{\partial t^2} &= -\varepsilon\mu_0 \frac{\partial}{\partial t} \left[ \frac{\partial \mathbf{E}_n}{\partial t} e^{i(k_n z - \omega_n t)} - \mathbf{E}_n i\omega_n e^{i(k_n z - \omega_n t)} \right] \\ &= -\varepsilon\mu_0 \left[ \frac{\partial^2 \mathbf{E}_n}{\partial t^2} e^{i(k_n z - \omega_n t)} - \frac{\partial \mathbf{E}_n}{\partial t} i\omega_n e^{i(k_n z - \omega_n t)} - \frac{\partial \mathbf{E}_n}{\partial t} i\omega_n e^{i(k_n z - \omega_n t)} - \mathbf{E}_n \omega_n^2 e^{i(k_n z - \omega_n t)} \right] \\ &= -\varepsilon\mu_0 \frac{\partial^2 \mathbf{E}_n}{\partial t^2} e^{i(k_n z - \omega_n t)} + 2i\omega_n \varepsilon\mu_0 \frac{\partial \mathbf{E}_n}{\partial t} e^{i(k_n z - \omega_n t)} + \varepsilon\mu_0 \omega_n^2 \mathbf{E}_n e^{i(k_n z - \omega_n t)} \end{aligned}$$


---

$$\frac{\partial^2 \mathbf{E}_n}{\partial z^2} + 2ik_n \frac{\partial \mathbf{E}_n}{\partial z} - \varepsilon\mu_0 \frac{\partial^2 \mathbf{E}_n}{\partial t^2} + 2i\omega_n \varepsilon\mu_0 \frac{\partial \mathbf{E}_n}{\partial t} = -\mu_0 \omega_n^2 \mathbf{P}_n^{NL} e^{i\Delta k z}$$

采用空间的慢变振幅近似

$$\frac{\partial^2 \mathbf{E}_n}{\partial z^2} + 2ik_n \frac{\partial \mathbf{E}_n}{\partial z} - \epsilon\mu_0 \frac{\partial^2 \mathbf{E}_n}{\partial t^2} + 2i\omega_n \epsilon\mu_0 \frac{\partial \mathbf{E}_n}{\partial t} = -\mu_0 \omega_n^2 \mathbf{P}_n^{NL} e^{i\Delta kz}$$

时间慢变近似:  $\left| \frac{\partial^2 \mathbf{E}_n}{\partial t^2} \right| \ll \left| \omega_n \frac{\partial \mathbf{E}_n}{\partial t} \right|$

$$\frac{\partial \mathbf{E}_n}{\partial z} + \frac{n}{c} \frac{\partial \mathbf{E}_n}{\partial t} = \frac{i\omega_n}{2\epsilon_0 cn} \mathbf{P}_n^{NL} e^{i\Delta kz}$$

渡越时间  $T = L_n/c$  内振幅变化不显著时, 可忽略方程中的时间导数。



## 4.2.2 光的二次谐波

非线性极化率  $\xrightarrow{\text{代入}}$  极化强度  $\xrightarrow{\text{边界条件}}$  非线性耦合波方程  $\longrightarrow$  结果

$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + c.c.$$

二阶非线性极化强度的表达式:

$$\begin{aligned} P^{(2)}(t) &= \varepsilon_0 \chi^{(2)} E(t)^2 \\ &= \varepsilon_0 \chi^{(2)} \left[ E_1^2 e^{-2i\omega_1 t} + E_2^2 e^{-2i\omega_2 t} + 2E_1 E_2 e^{-i(\omega_1 + \omega_2)t} + 2E_1 E_2^* e^{-i(\omega_1 - \omega_2)t} + c.c. \right] + 2\varepsilon_0 \chi^{(2)} \left[ E_1 E_1^* + E_2 E_2^* \right] \end{aligned}$$

$$P^{(2)}(t) = \sum_n P(\omega_n) e^{-i\omega_n t}$$

$$P(2\omega_1) = \varepsilon_0 \chi^{(2)} E_1^2$$

二次谐波产生(SHG)

$$P(2\omega_2) = \varepsilon_0 \chi^{(2)} E_2^2$$

二次谐波产生(SHG)

$$P(\omega_1 + \omega_2) = 2\varepsilon_0 \chi^{(2)} E_1 E_2$$

和频(SFG)

$$P(\omega_1 - \omega_2) = 2\varepsilon_0 \chi^{(2)} E_1 E_2^*$$

差频(DFG)

$$P(0) = 2\varepsilon_0 \chi^{(2)} (E_1 E_1^* + E_2 E_2^*)$$

光学整流(OR)

负频率的响应:

$$P(-2\omega_1) = \varepsilon_0 \chi^{(2)} E_1^{*2}$$

$$P(-2\omega_2) = \varepsilon_0 \chi^{(2)} E_2^{*2}$$

$$P(-\omega_1 - \omega_2) = 2\varepsilon_0 \chi^{(2)} E_1^* E_2^*$$

$$P(\omega_2 - \omega_1) = 2\varepsilon_0 \chi^{(2)} E_2 E_1^*$$

用实数表示的非线性过程：

$$E(t) = E_1 \cos \omega_1 t + E_2 \cos \omega_2 t$$

$$\begin{aligned} E^2(t) &= (E_1 \cos \omega_1 t + E_2 \cos \omega_2 t)^2 \\ &= E_1^2 \cos^2 \omega_1 t + E_2^2 \cos^2 \omega_2 t + 2E_1 E_2 \cos(\omega_1 t) \cos(\omega_2 t) \\ &= \frac{1}{2} E_1^2 (1 + \cos 2\omega_1 t) + \frac{1}{2} E_2^2 (1 + \cos 2\omega_2 t) \\ &\quad + E_1 E_2 [\cos(\omega_1 + \omega_2) t + \cos(\omega_1 - \omega_2) t] \end{aligned}$$

矢量表达式:  $\mathbf{P}^{(2)} = \epsilon_0 \chi^{(2)} : \mathbf{E} \mathbf{E}$

和频:  $\mathbf{P}^{(2)}(\omega_1 + \omega_2) = \epsilon_0 \chi^{(2)}(-\omega_1 - \omega_2, \omega_1, \omega_2) : \mathbf{E}(\omega_1) \mathbf{E}(\omega_2)$

## 三光波相互作用的耦合波方程

$$\frac{\partial \mathbf{E}_n}{\partial z} = \frac{i\omega_n}{2\varepsilon_0 cn} \mathbf{P}_n^{NL} e^{i\Delta k z}$$

非线性源项:  $\mathbf{P}^{(2)}(\omega_1) = 2\varepsilon_0 \chi^{(2)}(-\omega_1; \omega_3, -\omega_2) : \mathbf{E}_3 \mathbf{E}_2^*$

$\omega_3 = \omega_1 + \omega_2$   $\mathbf{P}^{(2)}(\omega_2) = 2\varepsilon_0 \chi^{(2)}(-\omega_2; \omega_3, -\omega_1) : \mathbf{E}_3 \mathbf{E}_1^*$

$\mathbf{P}^{(2)}(\omega_3) = 2\varepsilon_0 \chi^{(2)}(-\omega_3; \omega_1, \omega_2) : \mathbf{E}_1 \mathbf{E}_2$

忽略吸收，代入到非线性波动方程：

$$\frac{d\mathbf{E}_1(z)}{dz} = \frac{i\omega_1}{cn_1} \chi^{(2)}(-\omega_1; \omega_3, -\omega_2) : \mathbf{E}_3(z) \mathbf{E}_2^*(z) e^{i\Delta k z}$$

$$\frac{d\mathbf{E}_2(z)}{dz} = \frac{i\omega_2}{cn_2} \chi^{(2)}(-\omega_2; \omega_3, -\omega_1) : \mathbf{E}_3(z) \mathbf{E}_1^*(z) e^{i\Delta k z}$$

$$\frac{d\mathbf{E}_3(z)}{dz} = \frac{i\omega_3}{cn_3} \chi^{(2)}(-\omega_3; \omega_1, \omega_2) : \mathbf{E}_1(z) \mathbf{E}_2(z) e^{-i\Delta k z}$$

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

$\Delta k = 0$ : 相位匹配

引入光电场偏振方向的单位矢量 $\mathbf{e}$ :  $\mathbf{E}_i(z) = E_i(z)\mathbf{e}_i \quad i = 1, 2, 3$

$$\frac{d\mathbf{E}_1(z)}{dz} = \frac{i\omega_1}{cn_1} \chi^{(2)}(-\omega_1; \omega_3, -\omega_2) : \mathbf{E}_3(z) \mathbf{E}_2^*(z) e^{i\Delta kz}$$



$$\frac{dE_1(z)}{dz} \mathbf{e}_1 = \frac{i\omega_1}{cn_1} \chi^{(2)}(-\omega_1; \omega_3, -\omega_2) : E_3(z) \mathbf{e}_3 E_2^*(z) \mathbf{e}_2 e^{i\Delta kz}$$

$$\mathbf{e}_1 \cdot \frac{dE_1(z)}{dz} \mathbf{e}_1 = \mathbf{e}_1 \cdot \frac{i\omega_1}{cn_1} \chi^{(2)}(-\omega_1; \omega_3, -\omega_2) : E_3(z) \mathbf{e}_3 E_2^*(z) \mathbf{e}_2 e^{i\Delta kz}$$

$$\frac{dE_1(z)}{dz} = \frac{i\omega_1}{cn_1} \chi^{(2)}(-\omega_1; \omega_3, -\omega_2) E_3(z) E_2^*(z) e^{i\Delta kz}$$

$$\chi^{(2)}(-\omega_1; \omega_3, -\omega_2) = \mathbf{e}_1 \cdot \chi^{(2)}(-\omega_1; \omega_3, -\omega_2) : \mathbf{e}_3 \mathbf{e}_2$$

标量形式的耦合波方程：

$$\frac{dE_1(z)}{dz} = \frac{i\omega_1}{cn_1} \chi^{(2)}(-\omega_1; \omega_3, -\omega_2) E_3(z) E_2^*(z) e^{i\Delta kz}$$

$$\frac{dE_2(z)}{dz} = \frac{i\omega_2}{cn_2} \chi^{(2)}(-\omega_2; \omega_3, -\omega_1) E_3(z) E_1^*(z) e^{i\Delta kz}$$

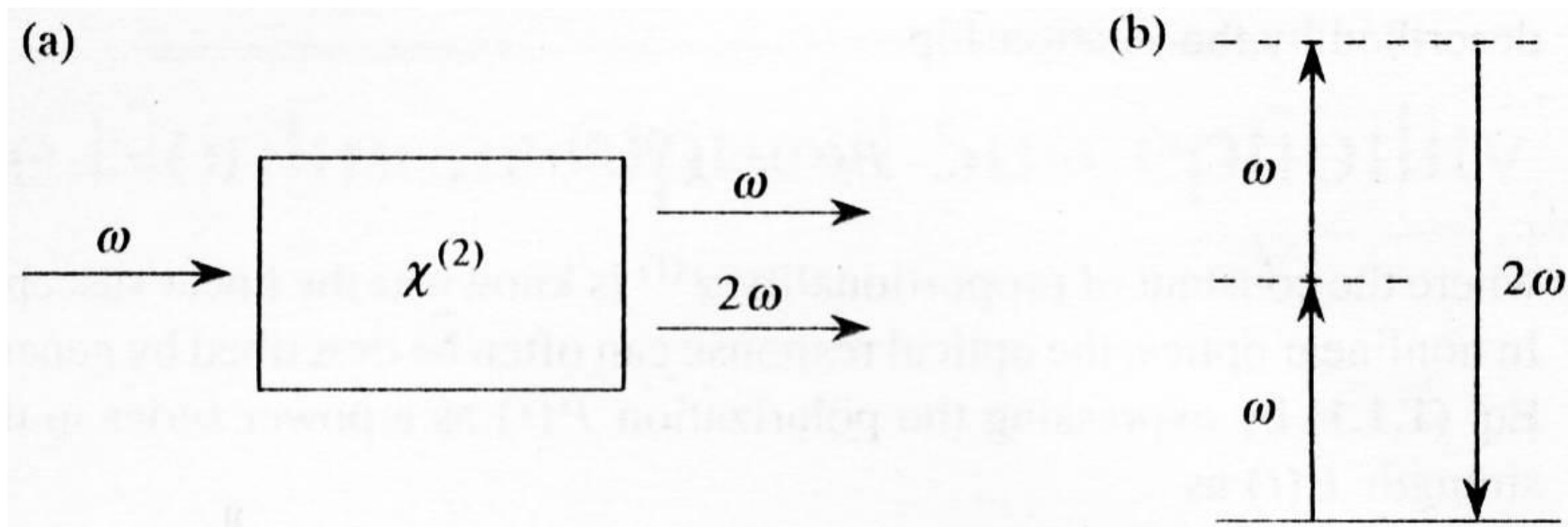
$$\frac{dE_3(z)}{dz} = \frac{i\omega_3}{cn_3} \chi^{(2)}(-\omega_3; \omega_1, \omega_2) E_1(z) E_2(z) e^{-i\Delta kz}$$

$$\chi^{(2)}(-\omega_1; \omega_3, -\omega_2) = \mathbf{e}_1 \cdot \boldsymbol{\chi}^{(2)}(-\omega_1; \omega_3, -\omega_2) : \mathbf{e}_3 \mathbf{e}_2$$

$$\chi^{(2)}(-\omega_2; \omega_3, -\omega_1) = \mathbf{e}_2 \cdot \boldsymbol{\chi}^{(2)}(-\omega_2; \omega_3, -\omega_1) : \mathbf{e}_3 \mathbf{e}_1$$

$$\chi^{(2)}(-\omega_3; \omega_1, \omega_2) = \mathbf{e}_3 \cdot \boldsymbol{\chi}^{(2)}(-\omega_3; \omega_1, \omega_2) : \mathbf{e}_1 \mathbf{e}_2$$

## 二次谐波产生



$$2\omega_1 = 2\omega_2 = \omega_3$$

对于小信号近似:  $E_3 \ll E_1$



$$\frac{dE_1(z)}{dz} = 0$$

$$\frac{dE_3(z)}{dz} = \frac{i\omega_3}{2cn_3} \chi^{(2)} E_1^2 e^{-i\Delta kz}$$

$$\Delta k = 2\omega [n(\omega) - n(2\omega)] / c$$

边界条件为  $E_3(0) = 0$

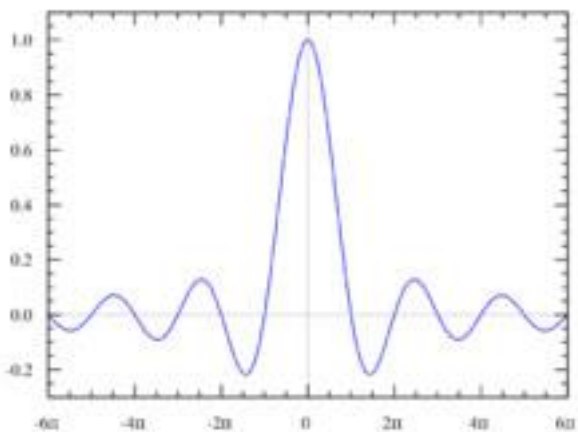
$$E_3(L) = -\frac{\omega_1}{cn_3\Delta k} \chi^{(2)} E_1^2(0)(e^{-i\Delta kL} - 1)$$

$$I = \frac{1}{2} \epsilon_0 cn |\mathbf{E}|^2$$

在介质出射面 $z = L$ ，倍频光的强度为：

$$I_3 = \frac{2\omega_1^2 [\chi^{(2)}]^2 I_1^2 L^2}{c^3 n_1^2 n_3 \epsilon_0} \cdot \frac{\sin^2(\Delta k L / 2)}{(\Delta k L / 2)^2} = \frac{2\omega_1^2 [\chi^{(2)}]^2 I_1^2 L^2}{c^3 n_1^2 n_3 \epsilon_0} \operatorname{sinc}^2\left(\frac{\Delta k L}{2}\right)$$

光倍频转换效率： $\eta = \frac{P_3}{P_1} = \frac{2\omega_1^2 [\chi^{(2)}]^2 L^2}{c^3 n_1^2 n_3 \epsilon_0} \cdot \frac{P_1}{A} \cdot \operatorname{sinc}^2\left(\frac{\Delta k L}{2}\right)$



sinc函数

$$(1) \quad I_3 \propto I_1^2$$

$$(2) \quad I_3 \propto \operatorname{sinc}^2\left(\frac{\Delta k L}{2}\right)$$

如果  $\Delta k = 0$ ,  $I_3 \propto L^2$

$$(3) \quad \eta \propto I_1$$

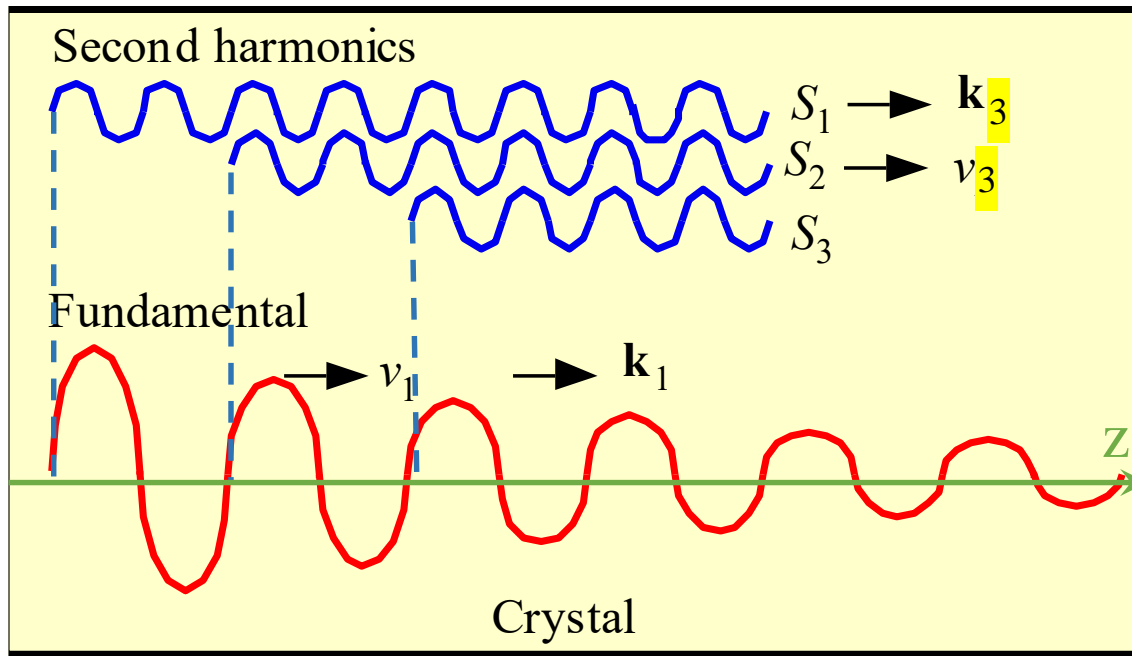
$$(4) \quad \eta \propto [\chi^{(2)}]^2$$

$$\Delta k = \frac{\omega n(\omega)}{c} + \frac{\omega n(\omega)}{c} - \frac{2\omega n(2\omega)}{c} = \frac{2\omega n(\omega)}{c} - \frac{2\omega n(2\omega)}{c}$$

$k_1$                        $k_2$                        $k_3$

$\uparrow$   
 $2\omega$ 的极化

$\uparrow$   
 $2\omega$ 的光



能量守恒和动量守恒的条件为：

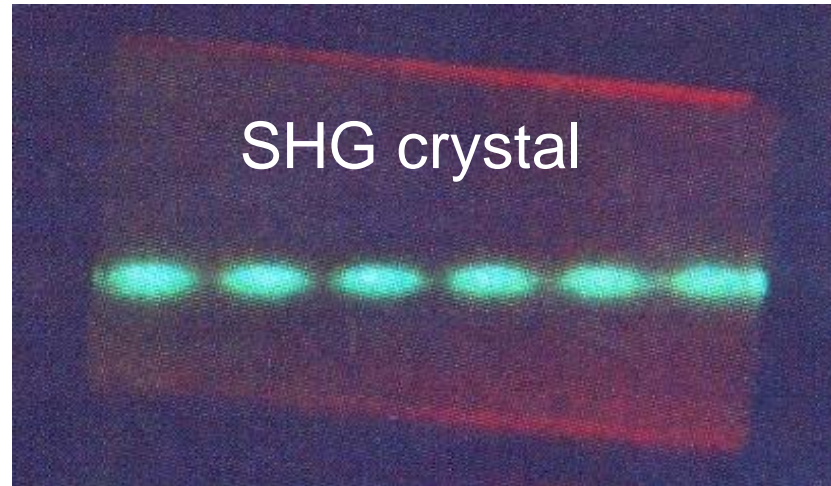
$$\begin{aligned}\omega_1 + \omega_2 &= \omega_3 \\ k_1 + k_2 &= k_3\end{aligned}$$

当 $v_p(\omega) = v_p(2\omega)$ , 即 $n(\omega) = n(2\omega)$ 时,  
空间每一点产生的倍频光同相相加, 输出最大。

# 晶体中的二次谐波产生

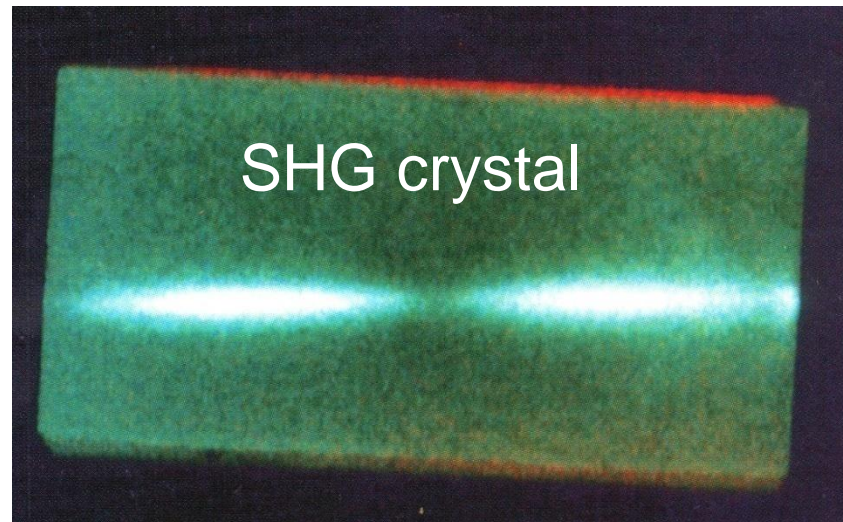
Far from  
phase-matching:

→  
Input beam

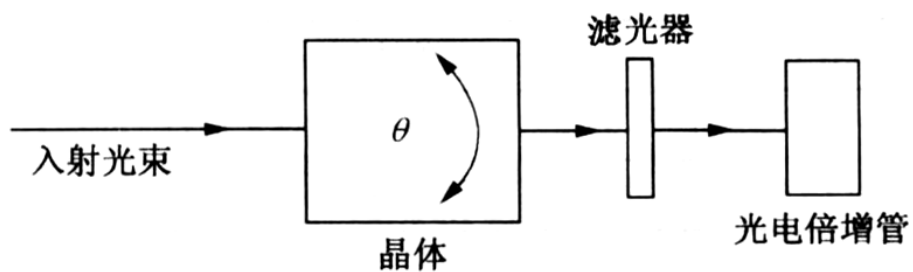


Closer to  
phase-matching:

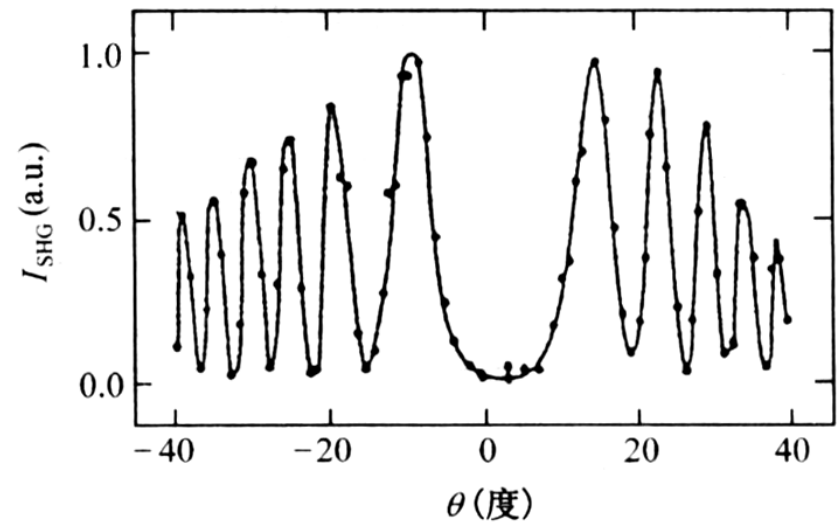
→  
Input beam



## 四. 相位匹配技术



(a) Maker条纹的实验装置



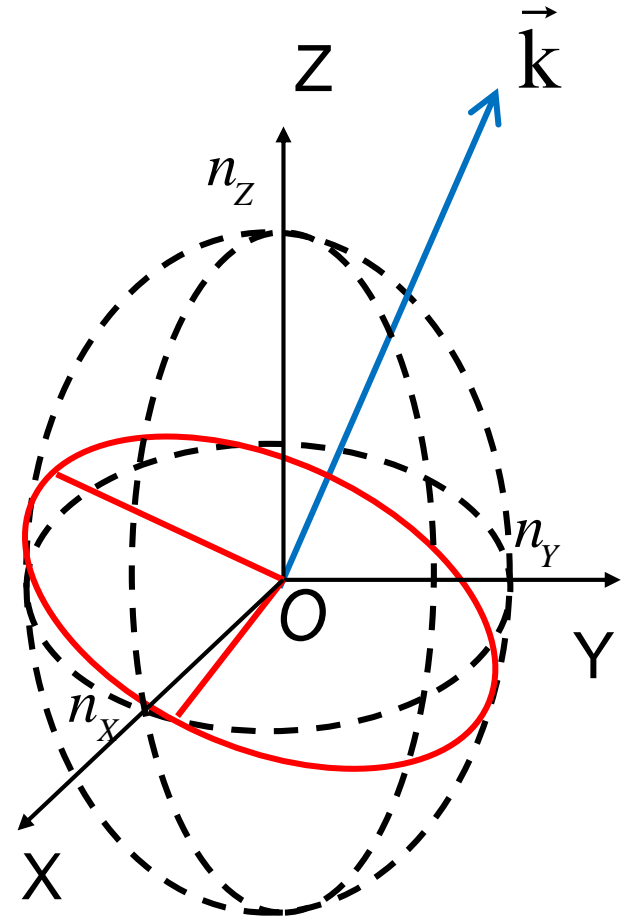
(b) 倍频光的Maker条纹

# 折射率椭球面

$$\frac{X^2}{n_X^2} + \frac{Y^2}{n_Y^2} + \frac{Z^2}{n_Z^2} = 1$$

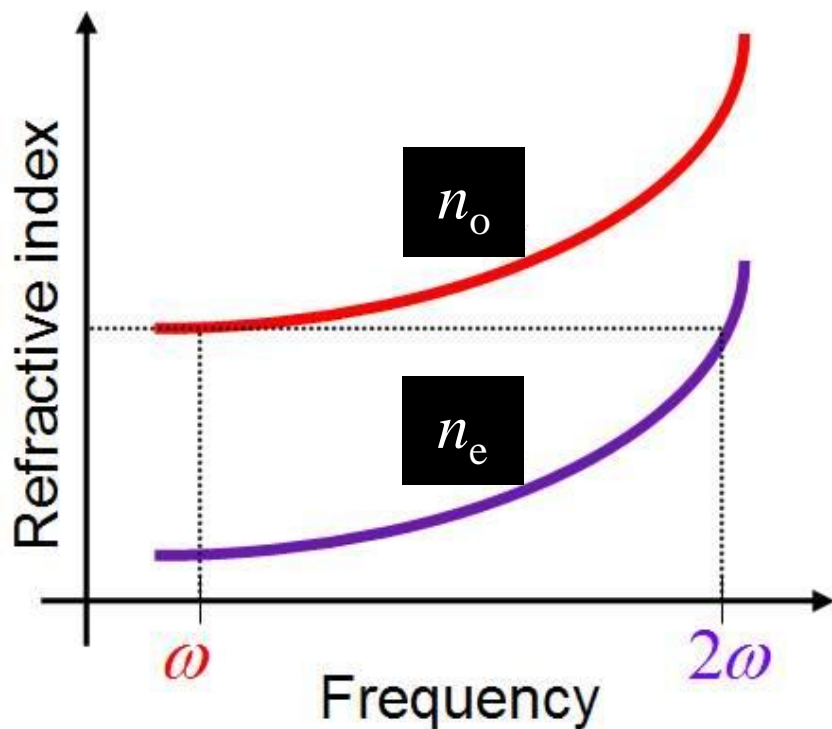
光在各向异性材料中的传播特性：

- ✓ 过原点作一与传播光波波矢垂直的平面
- ✓ 椭圆的长短轴方向为光的两个本征偏振方向，半轴长既是相应的两个折射率。

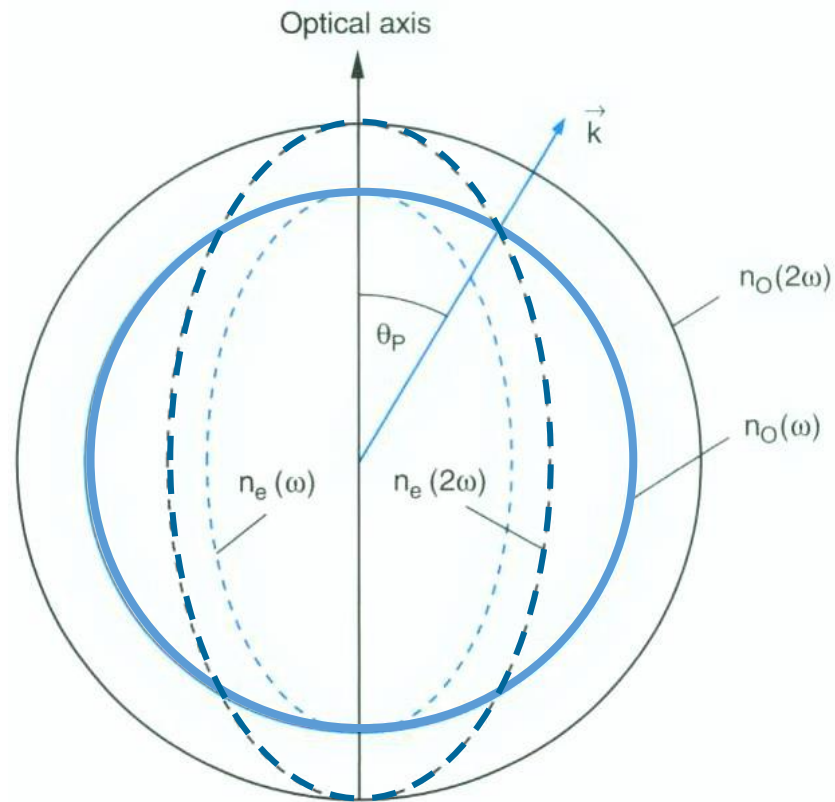


# 利用双折射达到相位匹配

寻常光 (o光) : 偏振垂直于入射方向和光轴形成的平面;  
非常光 (e光) : 偏振在入射方向和光轴形成的平面内;  
o光和e光的折射率差可达0.1



负单轴晶体



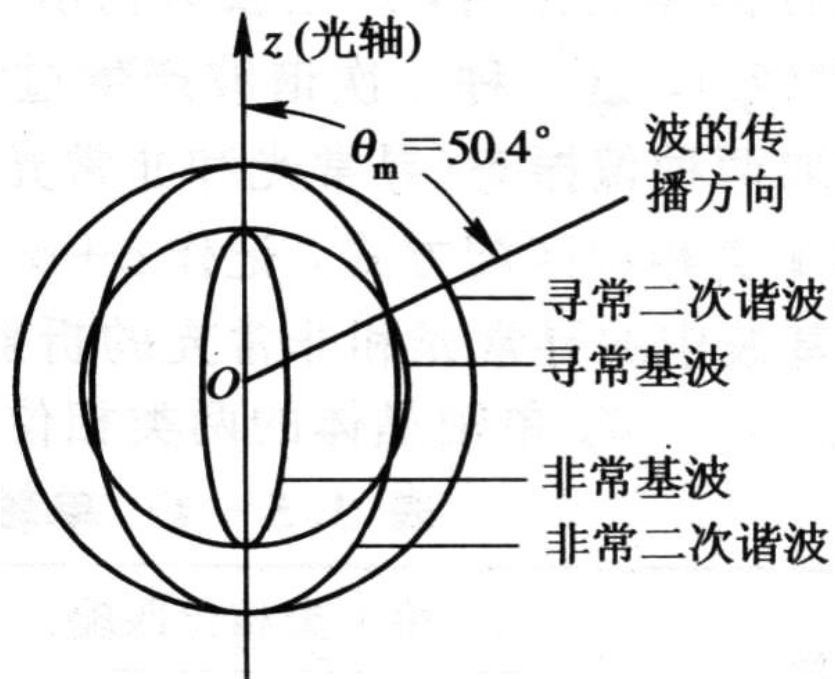
$n_e$ 依赖于入射角度

# (1) 角度相位匹配

表 4.1 单轴晶体的相位匹配条件

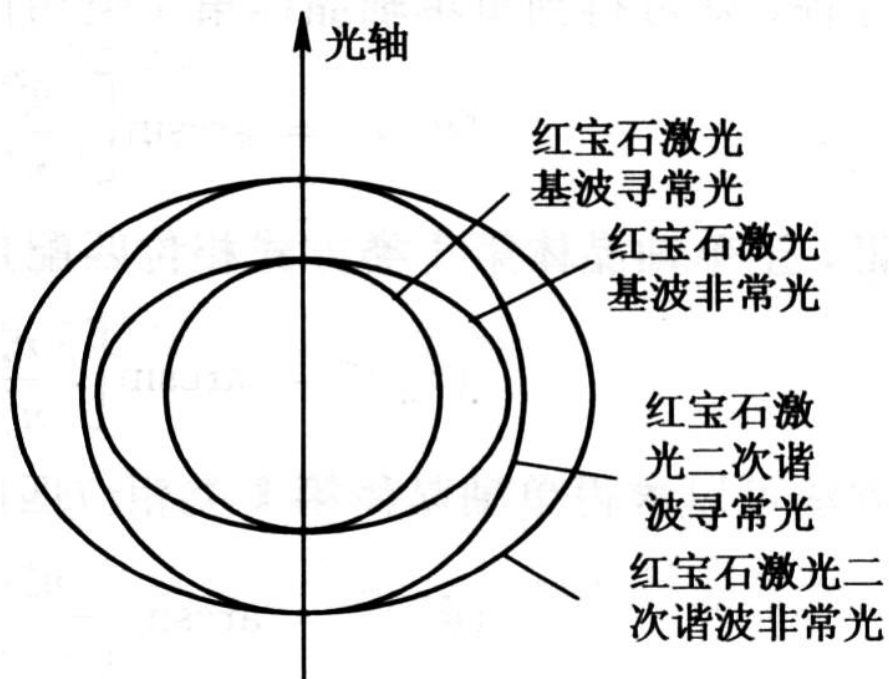
晶体种类	第 I 类相位匹配		第 II 类相位匹配	
	偏振性质	相位匹配条件	偏振性质	相位匹配条件
正单轴晶体 ( $n_e > n_o$ )	$e + e \rightarrow o$	$n_e^\omega(\theta_m) = n_o^{2\omega}$	$o + e \rightarrow o$	$\frac{1}{2}[n_o^\omega + n_e^\omega(\theta_m)] = n_o^{2\omega}$
负单轴晶体 ( $n_e < n_o$ )	$o + o \rightarrow e$	$n_o^\omega = n_e^{2\omega}(\theta_m)$	$e + o \rightarrow e$	$\frac{1}{2}[n_e^\omega(\theta_m) + n_o^\omega] = n_e^{2\omega}(\theta_m)$





KDP晶体

(负单轴晶体)

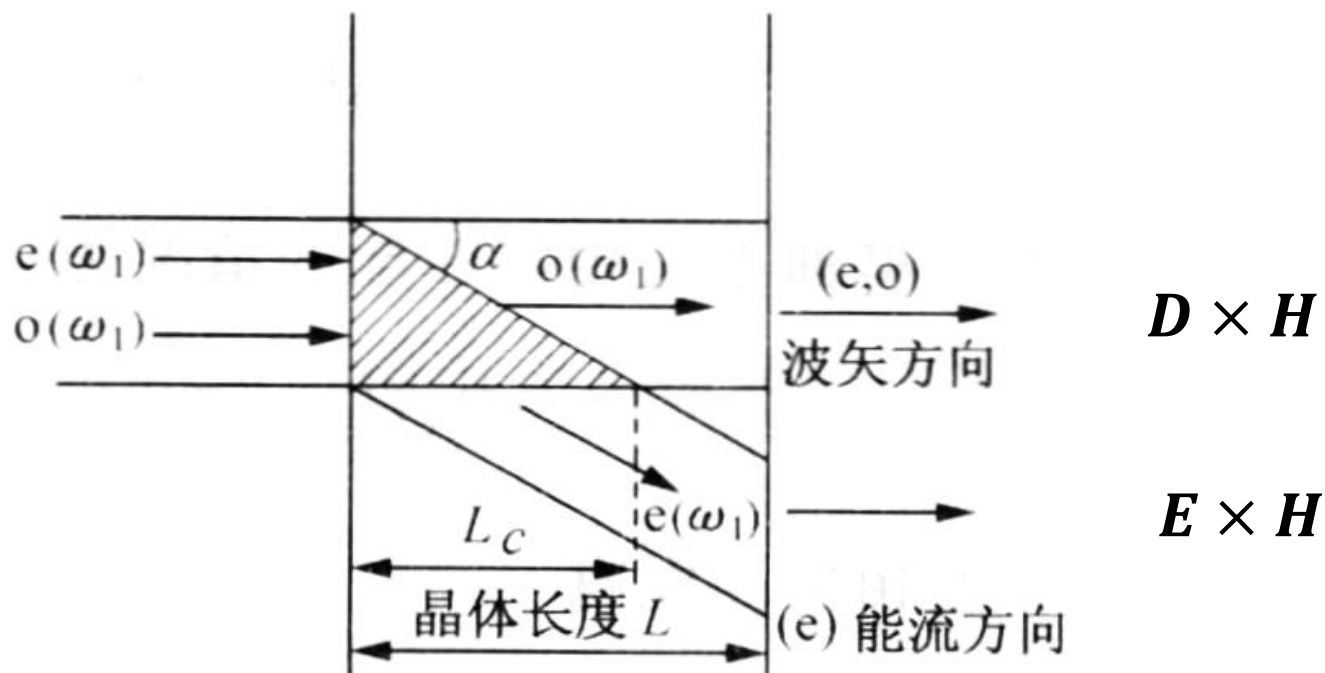


石英晶体

(正单轴晶体)

折射率曲线

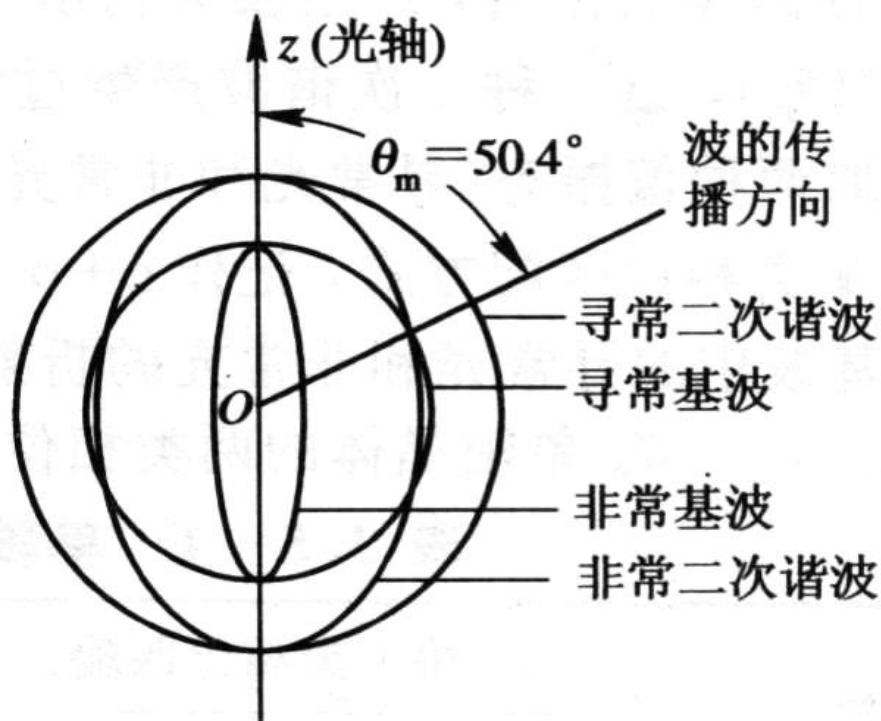
## (2) 温度匹配技术



双折射晶体中e光和o光的离散

为了不受离散效应的影响，必须使相位匹配角  $\theta_m = 90^\circ$

# 实现第I类相位匹配的负单轴晶体( $n_e < n_o$ ) 的温度匹配



$$\frac{\partial [n_o(2\omega) - n_o(\omega)]}{\partial T} > 0$$

正的色散温度特性

$$\frac{\partial [n_o(2\omega) - n_e(2\omega)]}{\partial T} < 0$$

负的双折射温度特性



相减

$$\frac{\partial [n_e(2\omega) - n_o(\omega)]}{\partial T} > 0$$

### (3) 相位匹配宽度

基于实际光束的情况，允许存在一个相位失配量。通常规定这一确定的相位失配量为

$$|\Delta k| = \pi / L$$

$L$ 为晶体的长度。在允许失配的最大处倍频转换效率降至完全匹配时的40%，但仍认为倍频有效。称这个确定的失配量为相位匹配宽度。

## 五. 常用的倍频晶体

对倍频晶体的要求：

- 大的二阶极化率
- 在工作波段有高的透过率
- 高损伤阈值。

实际应用中，还要求材料满足相位匹配的条件。根据基频光波长和晶体材料的具体数据（对称性、系数和折射率等），计算出晶体材料的切割角度，从而满足相位匹配的要求。

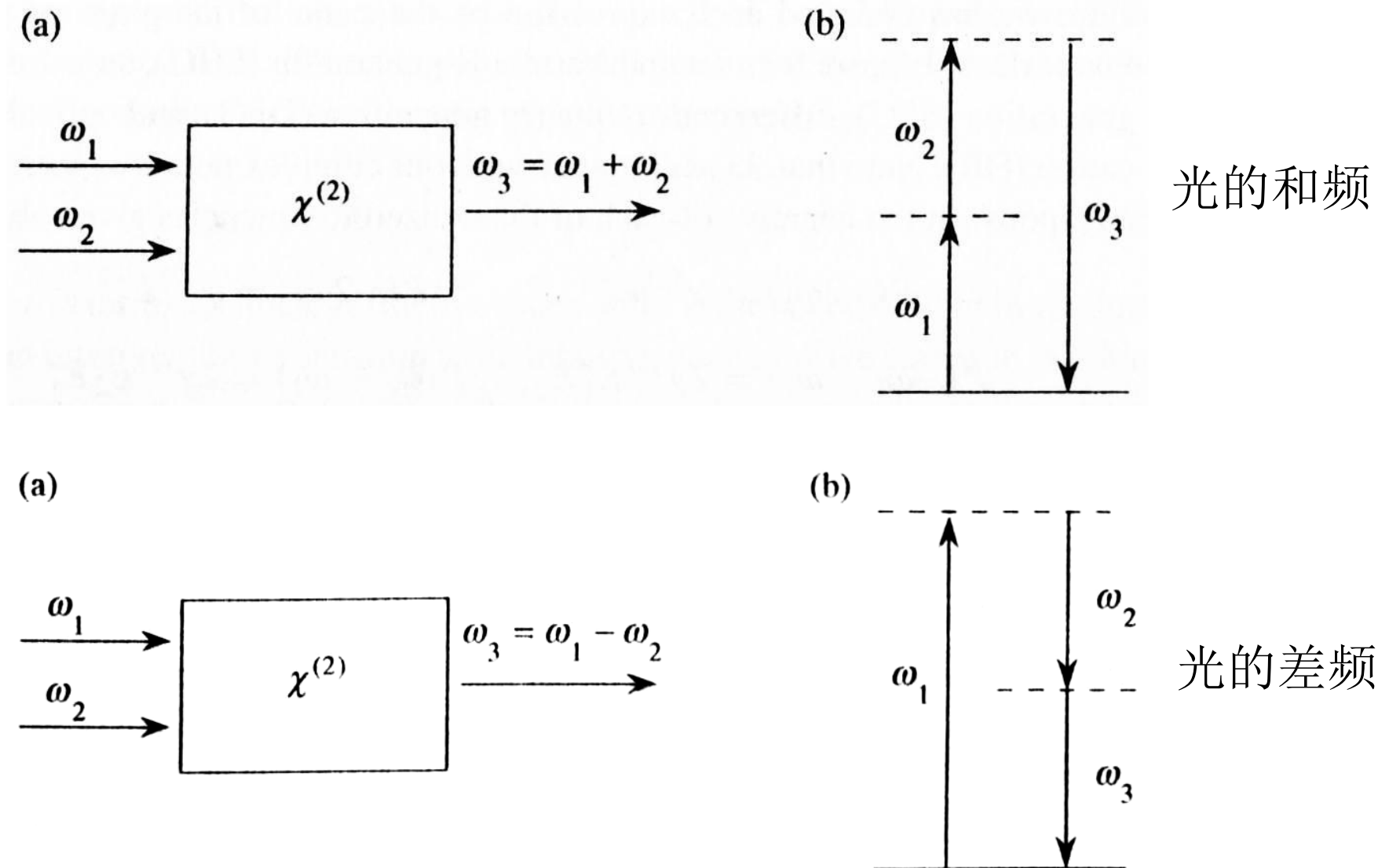
# 重要的非线性晶体

Material	Phase matching	Nonlinear. coeff. ( $10^{-12} \text{ mV}^{-1}$ )	$n_0$	Damage Threshold ( $\text{GWcm}^{-2}$ )
KDP	II	0.37	1.49	0.5
KTP	II	3.18	1.74	0.5
LBO	I	1.16	1.56	2.5
BBO	I	1.94	1.65	1.5
$\text{LiIO}_3$	I	4.1	1.85	0.01

KDP = potassium dihydrogen phosphate =  $\text{KH}_2\text{PO}_4$

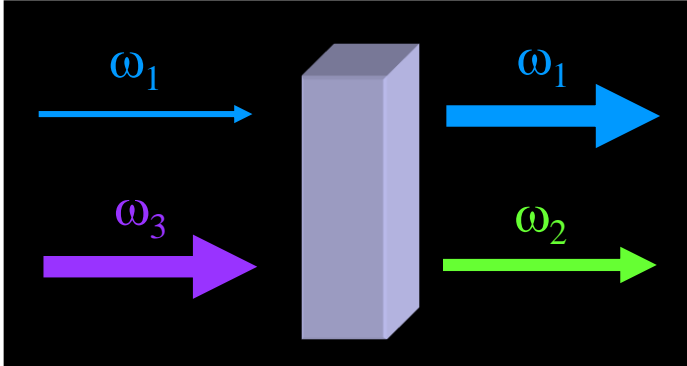
BBO = beta barium borate

### 4.3.3 其它二阶非线性光学现象

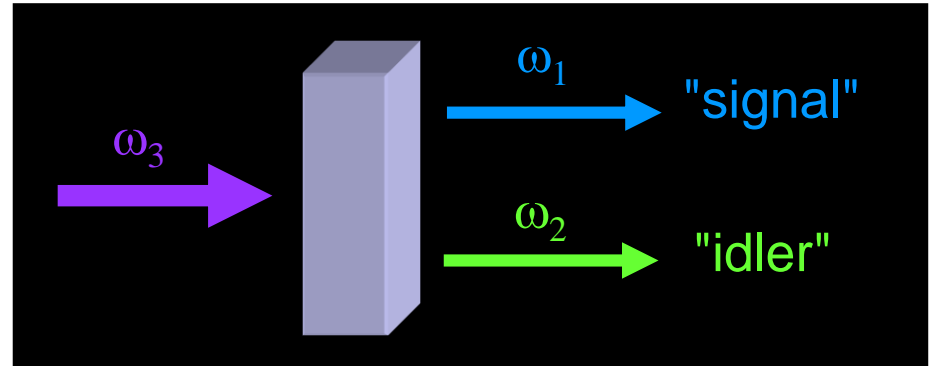


# 光学参量产生、放大、振荡

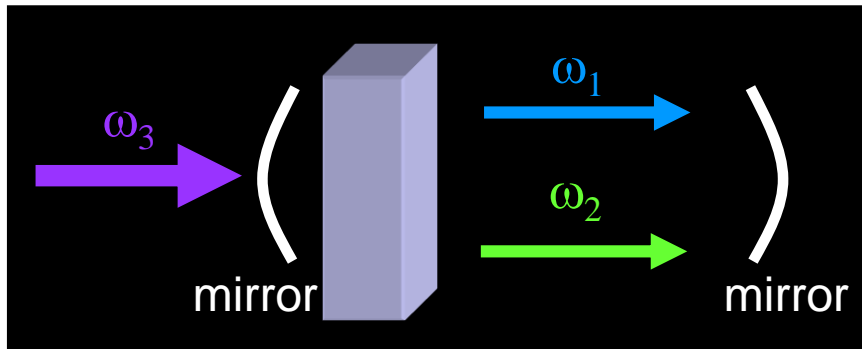
$$\omega_3 - \omega_1 = \omega_2$$



Optical Parametric Amplification (OPA)



Optical Parametric Generation (OPG)

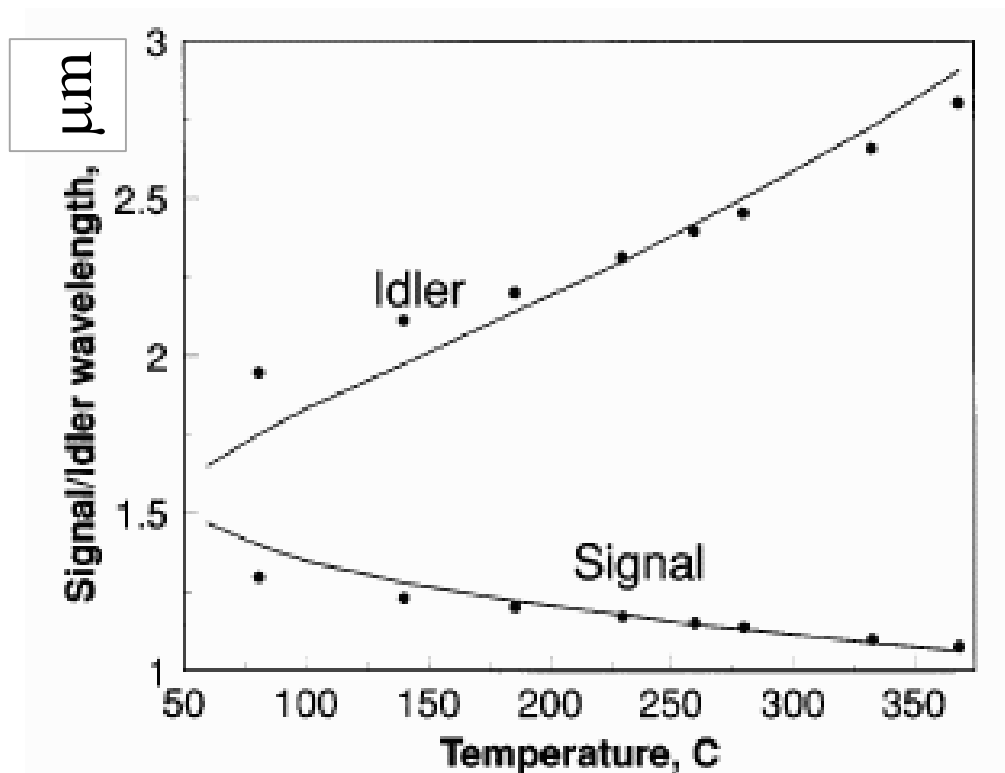


Optical Parametric Oscillation (OPO)



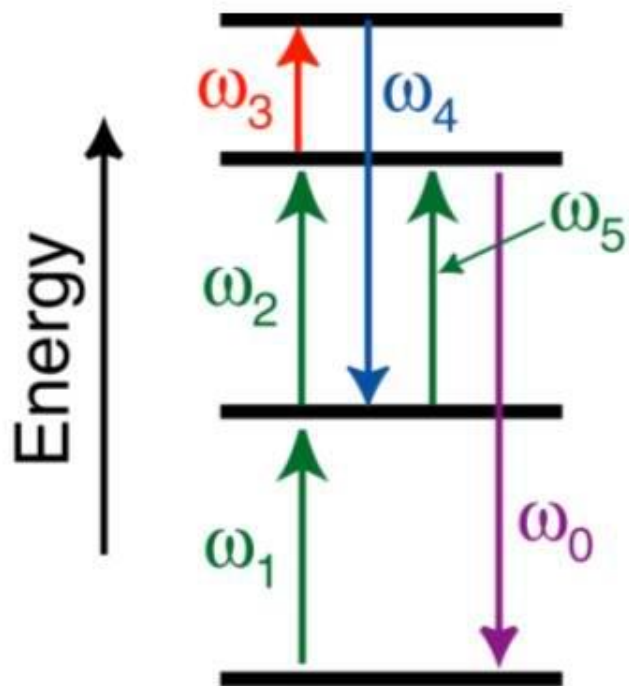
参量过程调谐输出波长的方法：

1. 改变晶体的角度；
2. 改变温度。

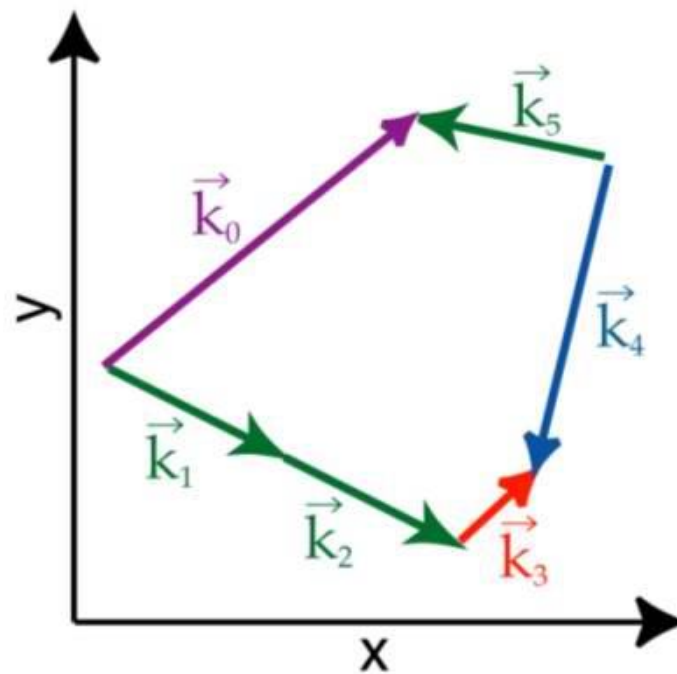


信号光和闲频光随温度的变化

## 4.4 三阶及高阶非线性光学过程

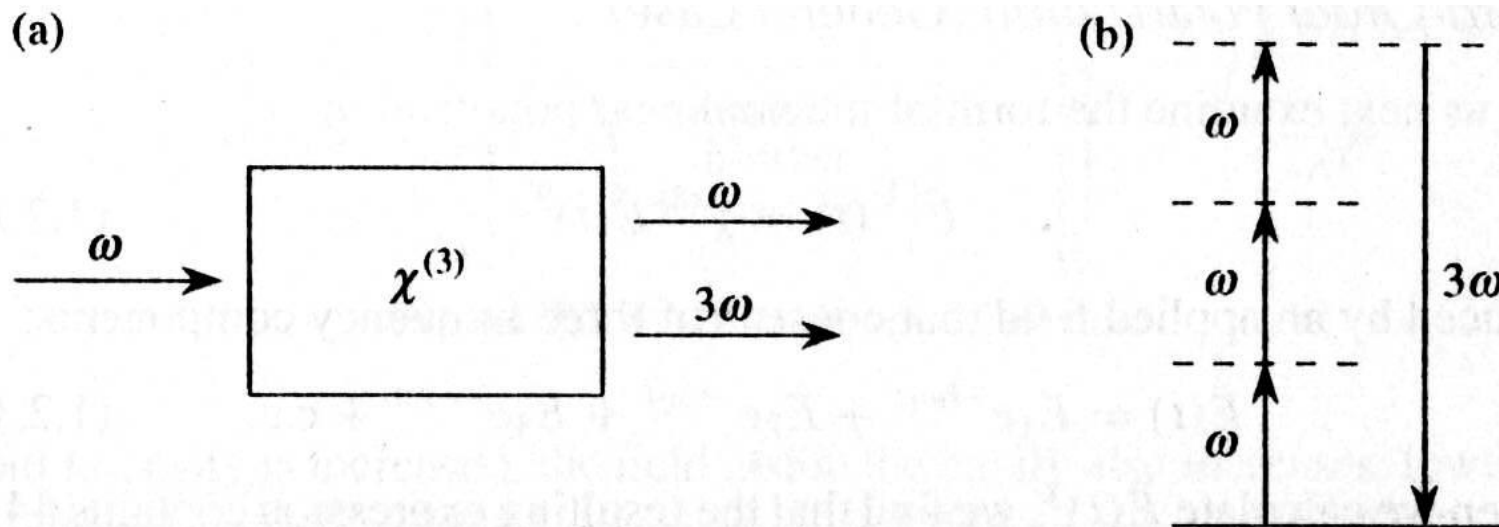


能量守恒



动量守恒

## 4.4.1 三次谐波产生



$$\mathbf{P}^{(3)} = \varepsilon_0 \chi^{(3)} : \mathbf{EEE}$$

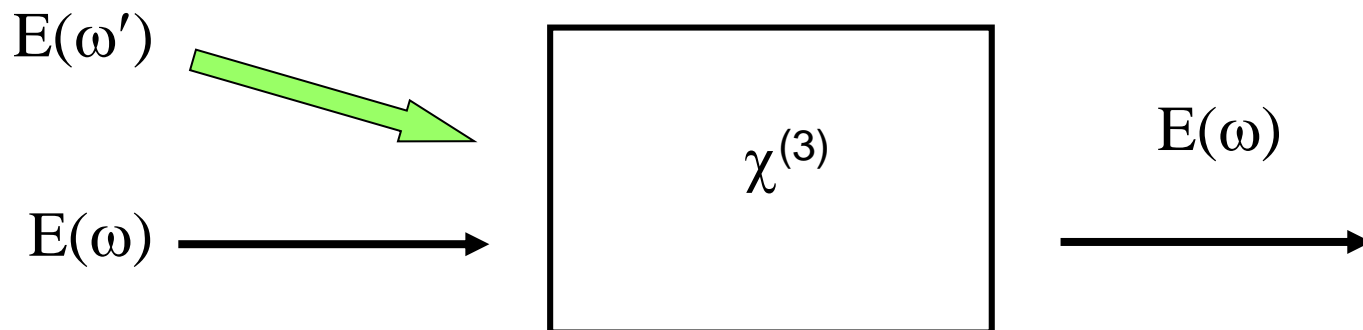
工作气体某一频率处在吸收：  $n_1(3\omega) < n_1(\omega)$

$$An_1(3\omega) + Bn_2(3\omega) = An_1(\omega) + Bn_2(\omega)$$

惰性气体无吸收：  $n_2(3\omega) > n_2(\omega)$

## 4.4.2 光克尔效应

光学克尔效应 —— 外加光场引起双折射的现象



$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P} = \varepsilon_r \varepsilon_0 \mathbf{E} \qquad \varepsilon_r = n^2 = 1 + \frac{|\mathbf{P}|}{\varepsilon_0 |\mathbf{E}|}$$

信号光:  $\mathbf{E}(\omega)$ , 单位方向矢量  $\mathbf{a}(\omega)$

泵浦光:  $\mathbf{E}(\omega')$ , 单位方向矢量  $\mathbf{a}(\omega')$

$$\mathbf{P}(\omega) = \mathbf{P}^{(1)}(\omega) + \mathbf{P}^{(3)}(\omega)$$

$$= \varepsilon_0 \left[ \chi^{(1)}(\omega) \cdot \mathbf{a}(\omega) + 6\chi^{(3)}(-\omega; \omega, -\omega', \omega') : \mathbf{a}(\omega) \mathbf{a}(\omega') \mathbf{a}(\omega') |E(\omega')|^2 \right] E(\omega)$$

克尔系数:  $K_{\omega'}(\omega) = \frac{\Delta n_{\parallel}(\omega) - \Delta n_{\perp}(\omega)}{\lambda |E(\omega')|^2}$

||/⊥: 信号光与泵浦光  
偏振平行/垂直

设泵浦光偏振沿y方向:  $\mathbf{a}(\omega') = (0, 1, 0)$

$$P_{\mu}^{(3)} = 6\varepsilon_0 E(\omega) |E(\omega')|^2 \sum_{\alpha=x,y,z} \chi_{\mu\alpha yy}^{(3)}(-\omega; \omega, -\omega', \omega') \underline{a_{\alpha}(\omega)}$$

$\alpha$ 方向投影

## 各向同性介质三阶极化率张量的非零元素

$$\chi_{xxxx}^{(3)} = \chi_{yyyy}^{(3)} = \chi_{zzzz}^{(3)}$$

$$\chi_{yyxx}^{(3)} = \chi_{zzyy}^{(3)} = \chi_{zzxx}^{(3)} = \chi_{xxzz}^{(3)} = \chi_{xxyy}^{(3)} = \chi_{yyxx}^{(3)}$$

$$\chi_{yzyz}^{(3)} = \chi_{zyzy}^{(3)} = \chi_{zxzx}^{(3)} = \chi_{xzxz}^{(3)} = \chi_{xyxy}^{(3)} = \chi_{yxyx}^{(3)}$$

$$\chi_{yzzx}^{(3)} = \chi_{zyyz}^{(3)} = \chi_{zxzx}^{(3)} = \chi_{xzzx}^{(3)} = \chi_{xyyx}^{(3)} = \chi_{yxyx}^{(3)}$$

并有

$$\chi_{xxxx}^{(3)} = \chi_{xxyy}^{(3)} + \chi_{xyxy}^{(3)} + \chi_{xyyx}^{(3)}$$

$$P_x^{(3)} = 6\varepsilon_0 \chi_{xxyy}^{(3)}(-\omega; \omega, -\omega', \omega') a_x(\omega) E(\omega) |E(\omega')|^2$$

$$P_y^{(3)} = 6\varepsilon_0 \chi_{yyyy}^{(3)}(-\omega; \omega, -\omega', \omega') a_y(\omega) E(\omega) |E(\omega')|^2$$

把上式代入耦合波方程

$$\frac{\partial E_y(\omega, z)}{\partial z} = \frac{3i\omega}{cn_\omega} \chi_{yyyy}^{(3)}(-\omega; \omega, -\omega', \omega') a_y(\omega) E(\omega) |E(\omega')|^2$$

$E(\omega')$ 不变:

$$E_y(\omega, z) \propto \exp \left\{ \frac{i\omega}{c} \left[ \frac{3\omega}{kc} |E(\omega')|^2 \chi_{yyyy}^{(3)}(-\omega; \omega, -\omega', \omega') \right] z \right\}$$

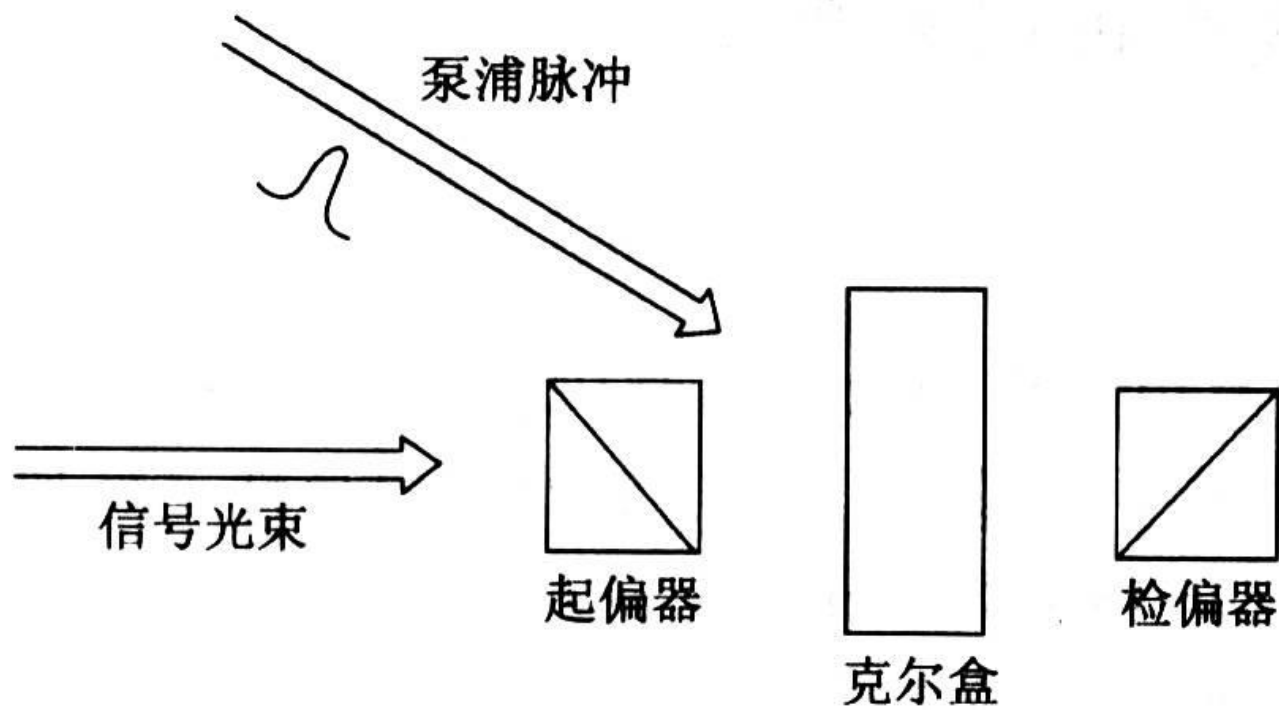
$$\Delta n_{\parallel} = \frac{3\omega}{kc} |E(\omega')|^2 \chi_{yyyy}^{(3)}(-\omega; \omega, -\omega', \omega')$$

$$\Delta n_{\perp} = \frac{3\omega}{kc} |E(\omega')|^2 \chi_{xxyy}^{(3)}(-\omega; \omega, -\omega', \omega')$$

$$K_{\omega'}(\omega) = \frac{\Delta n_{\parallel}(\omega) - \Delta n_{\perp}(\omega)}{\lambda |E(\omega')|^2}$$

$$K_{\omega'}(\omega) = \frac{3\omega}{2\pi c} \left[ \chi_{yyyy}^{(3)} - \chi_{xxyy}^{(3)} \right]$$

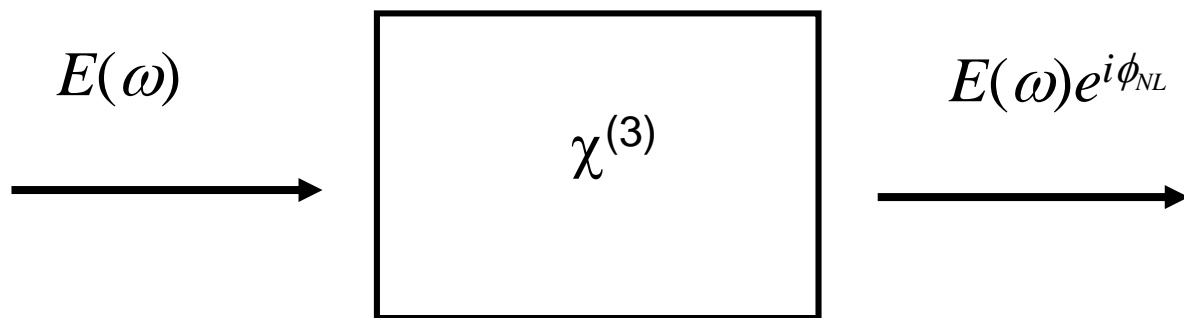




光克尔效应实验示意图

### 4.4.3 光的自聚焦

自聚焦 —— 折射率随光强变化而使光束在传播过程中出现聚焦



$$P(\omega) = P^{(1)}(\omega) + P^{(3)}(\omega) = \varepsilon_0 \chi^{(1)}(\omega) E(\omega) + 3\varepsilon_0 \chi^{(3)}(-\omega; \omega, -\omega, \omega) |E(\omega)|^2 E(\omega)$$

相对介电常数:  $\varepsilon_r = 1 + \chi^{(1)} + 3\chi^{(3)} |E(\omega)|^2$

$$n = \sqrt{\varepsilon_r} = \sqrt{1 + \chi^{(1)} + 3\chi^{(3)} |E(\omega)|^2} = n_0 + \Delta n$$

$$n_0 = \sqrt{1 + \chi^{(1)}}$$

线性折射率

非线性折射率

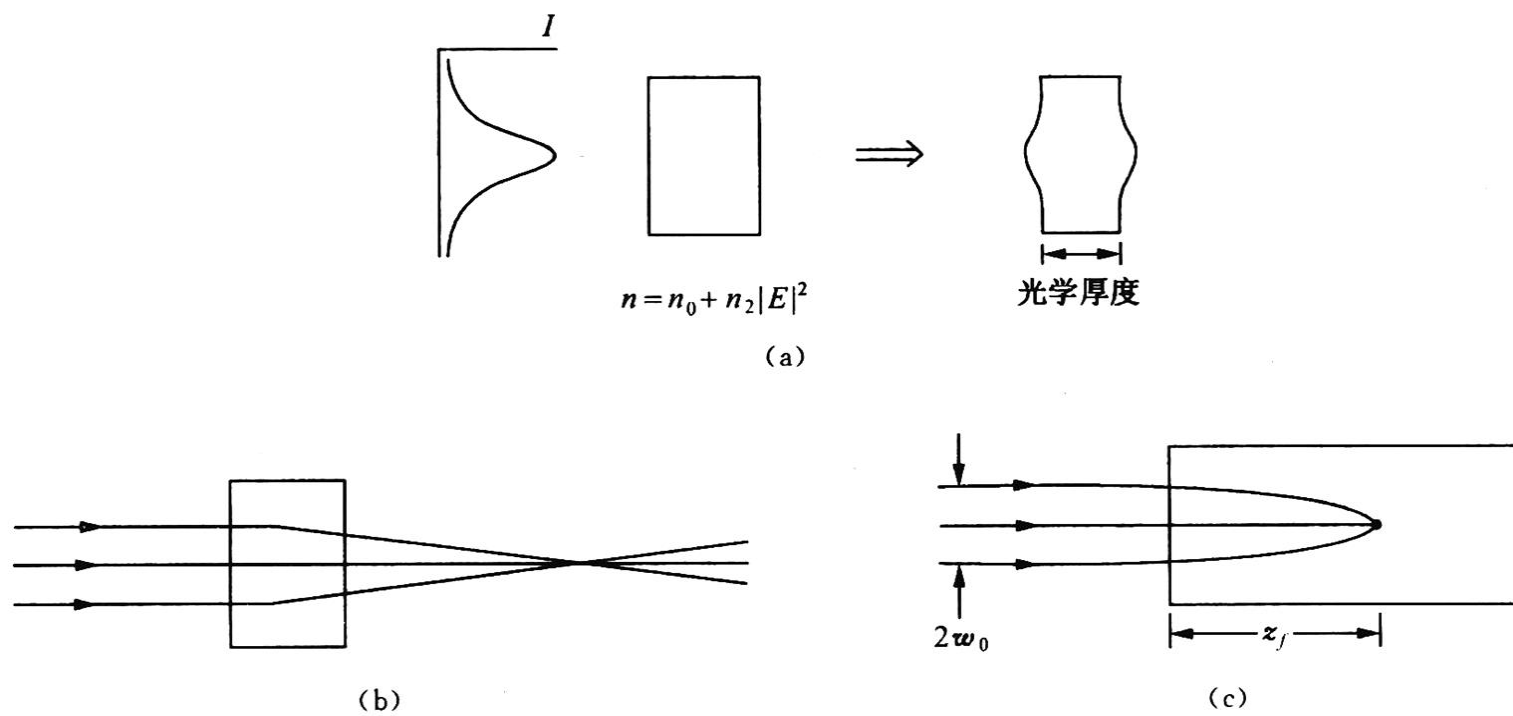
$$n = \sqrt{n_0^2 + 3\chi^{(3)} |E(\omega)|^2} = n_0 \sqrt{1 + \frac{3\chi^{(3)}}{n_0^2} |E(\omega)|^2}$$

通常  $n_0 \gg \Delta n$ :

$$n \approx n_0 \left( 1 + \frac{3\chi^{(3)}}{2n_0^2} |E(\omega)|^2 \right)$$

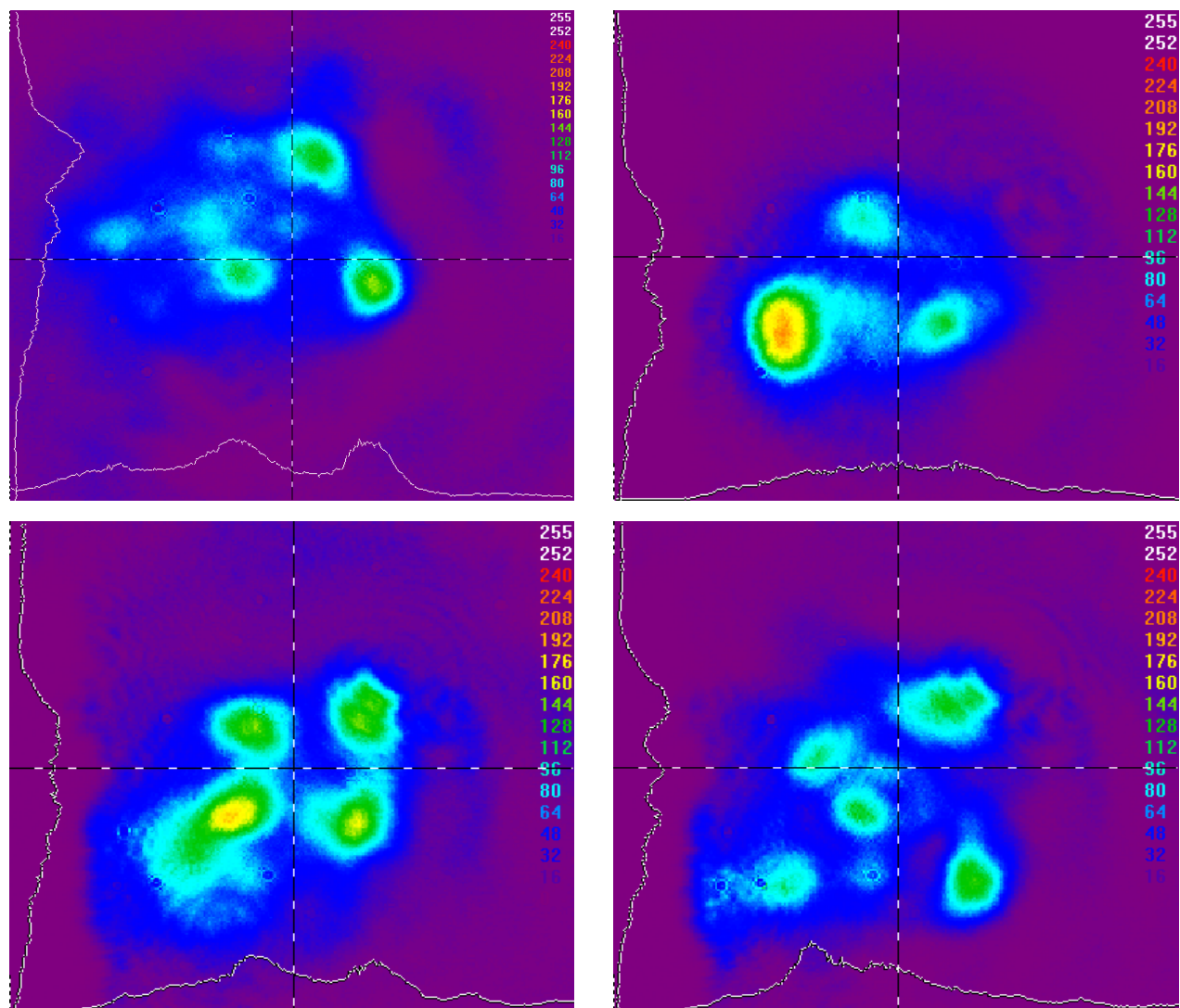
$$n_2 = \frac{3\chi^{(3)}}{2n_0} \quad n_2 \text{称为非线性折射率系数}$$

$$n = n_0 + n_2 |E(\omega)|^2$$




光的自聚焦示意图

# 强光束传输中的成丝现象



# 自相位调制


$$n(I) = n_0 + n_2 |E(\omega)|^2$$

Pulse intensity vs. time

$$E(z, t) = E(0, t) \exp \left[ i \frac{\omega}{c} n_2 |E(0, t)|^2 z \right]$$

传播越远，调制越大

$$\phi_{NL}(t) \approx \frac{\omega}{c} z n_2 |E(t)|^2$$

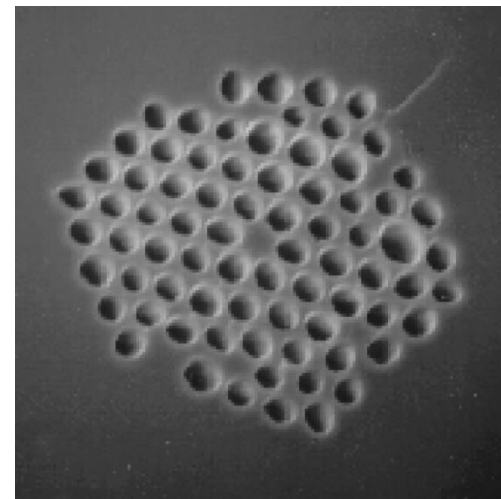
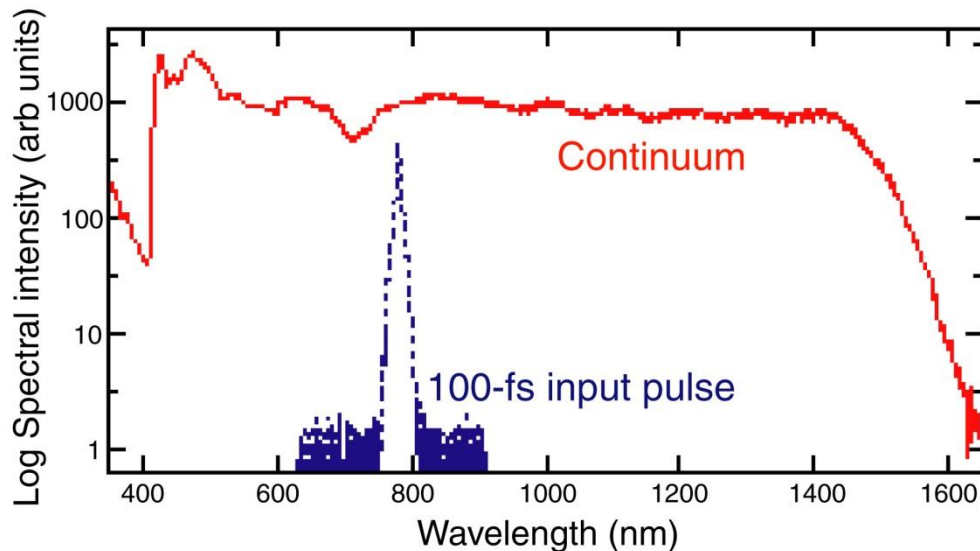
相位变化量

$$\Delta\omega_{inst}(t) = -\frac{\partial\phi_{NL}(t)}{\partial t} = -\frac{\omega z n_2}{c} \frac{\partial |E(t)|^2}{\partial t}$$

产生新的频率

对于超短激光脉冲，效应可能非常明显

# 利用光子晶体光纤产生超连续辐射



Cross section of the microstructure fiber.

- The spectrum extends from ~400 to ~1500 nm and is relatively flat (when averaged over time).

This continuum was created using *unamplified* Ti:Sapphire pulses.

J.K. Ranka, R.S. Windeler, and A.J. Stentz, Opt. Lett. Vol. 25, pp. 25-27, 2000



Continuum is quite beautiful!



# 非线性光学效应的应用

- 二阶非线性效应：倍频、和频、差频

转换效率高、广泛用于产生新的频率的激光

- 三阶非线性效应：光克尔、自聚焦、相位共轭、自相位调制

- 高阶非线性效应：高次谐波