6

a. $D(\sigma_v) = \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$ NON-zero 11, 22, 33, 21, 12

$D_{22} = 1$: this means we need an even number of "z" or y

$\Rightarrow$ xx; yy; zz are non-zero

b $D(C_4) = \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}$ $D_{21} = -1$
$D_{12} = D_{33} = 1$

$\Rightarrow$ D_{21} is going to bring down the number of non-zero/different elements

$\chi_{22}^{(1)'} = D_{21} D_{21} \cdot \chi_{11}^{(1)}$ must be true
$\chi_{11}^{(1)'} = D_{12} D_{12} \chi_{22}^{(1)}$ "

$\left.\begin{matrix} 11 = 22 ; \\ 22 = 11 ; \end{matrix}\right\}$ xx = yy

c. The same thing now applies also to the pairs xx, zz and yy, zz
So that xx = yy = zz $\Rightarrow$ just one value for $\chi^{(1)}$?

7a

KDP

$$D(C_2) = \underbrace{\begin{pmatrix} -1 & & 0 \\ & -1 & \\ 0 & & 1 \end{pmatrix}}$$

even numbers of (1,2) are necessary:

$\left\{ \begin{array}{l} 113,\ 223 + \text{permutations} \\ 123,\ 132 + \text{permutations} \\ 333 \end{array} \right.$

$$D(\sigma_d) = \underbrace{\begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}}$$

does not limit C_2 more than S_4 already did

$123 \to 213$

$$D(S_{4z}) = \underbrace{\begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}}$$

$113 \to -223$

$123 \to 213$

$312 \to 321$

$132 \to 231$

$$\chi_{113} = D_{\underline{2}1} D_{\underline{1}1} D_{\underline{3}3} \chi_{223}$$

this limits the selection done by $C_2(z)$ – we cross them out

$\left\{ \begin{array}{l} \cancel{113},\ \cancel{223} + \cancel{\text{permutations}} \\ 123,\ 132 + \text{permutations} \\ \cancel{333} \end{array} \right.$

$\Rightarrow$ final result for KDP

$$\boxed{\begin{array}{l} xyz = yxz \\ zxy = zyx \\ xzy = yzx \end{array}}$$

Another way of solving it:

$\chi_{123}, \chi_{213}, \chi_{312}, \chi_{321}, \chi_{132}, \chi_{213}$ are non-zero because we need ~~a combination of xyz that gives +1 when multiplying the matrix~~ elements in $D(S_4), D(C_2), D(\sigma_d)$

e.g. $\chi^{(2)}_{123} = D_{12} D_{21} D_{33} \chi^{(2)}_{213}$

$\begin{array}{cccc} -1 & +1 & -1 & (S_4) \\ +1 & +1 & +1 & (\sigma_d) \end{array}$

7 b

Cubic crystal - for Q (432)

$D(C_4)_x = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & -1 & 0 \end{pmatrix}$ $D(C_4)_y = \begin{pmatrix} 0 & 0 & -1 \\ 0 & 1 & 0 \\ 1 & 0 & 0 \end{pmatrix}$ $D(C_4)_z = \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}$

$D_{11} = D_{23} = 1$, $D_{32} = -1$

$D_{13} = -1$, $D_{22} = D_{31} = 1$

$D_{21} = -1$, $D_{12} = D_{33} = 1$

$\chi^{(2)\prime}_{lmn} = D_{il} \cdot D_{jm} \cdot D_{kn} \cdot \chi^{(2)}_{ijk}$

$C_{4,x}$

11	11	11	→ 111 = 111	
23	23	23	222 = 333	] 0, 111 also vanishes because of $C_{4,y}$ → xxx = yyy = zzz = 0
32	32	32	333 = −222	
11	23	11	121 = 131	] 0, xyx = xzx = 0; $C_{4,y}$ $C_{4,z}$!
11	32	11	131 = −121	yxy = yzy = 0
11	23	23	122 = 133	zxz = zyz = 0
11	32	32	133 = 122	
23	11	23	212 = 313	

even 23 no effect

xyy = xzz	yxy = zxz	yyx = zzx
yzz = yxx	zyz = xyx	zzy = xxy
zxx = ~~z~~ zyy	xzx = yzy	xxz = yyz

~~yyz = yyz~~

$C_{4,x}$

23	23	32	223 = −332	
32	32	23	332 = 223	yyz = −zzy = 0
23	32	23	yzy = − zyz	
32	23	32	232 = −323	] 0 yzy = zyz = 0
			323 = 232	
			zyz = yzy	
23	32	32	233 = 322	
32	23	23	322 = − 233	yzz = zyy = −yzz = 0

all 0

$C_{4,y}$: z,x pairs $C_{4,z}$ x,y pairs that are even are all 0

11	23	32	123 = −132	] xyz and all permutations are ~~allowed~~ allowed
11	32	23	132 = −123	

xyz = −xzy ; yxz = −zxy ; yzx = −zyx

⇒ anything with an even pair of x, y or z must vanish i.e. all possible tensor elements of ~~xyz~~ the type xxx, x−x, −xx, xx.. and permutations are zero.

$\underbrace{xyz = -xzy}$, $\overbrace{yxz = -zxy}^{C_{4,x,y}}$, $\underbrace{yzx = -zyx}_{C_{4,z...}}$ with $C_{4,x}$, $C_{4,y}$, $C_{4,z}$? →

$D(C_4)_z$

$$\chi^{(2)\prime}_{lmn} = D_{il} D_{jm} D_{kn} \chi^{(2)}_{ijh}$$

	33 12 21	312 = −321	zxy = −zyx
	33 21 12	321 = −312	
$D(C_4)_y$	22 13 31	213 = −231	yxz = −yzx
	22 31 13	231 = −213	
	12 33 21	132 = −231	xzy = −yzx
	21 33 12	231 = 132	

So we have: xyz = −xzy = yzx = −yxz = zxy = −zyx are non-zero.

To have these elements vanish we need either a σ or an i operation to make everything vanish.

c) With an i operation we have always −1 and no even x, y, z are available.

An isotropic medium has both $C_4 \times 3$ and i or σ and so all $\chi^{(2)}$ tensore elements vanish.

7d. For a surface we can assume an infinitesimal thin slab of nonlinear dipoles as source term.

$P^{(2)}$ has units $\frac{C \cdot m}{m^2}$. $\chi^{(2)}$ will also arise only from this slab.

<u>Bulk</u> $P^{(2)} = \epsilon_0 \chi^{(2)} \cdot E^2$

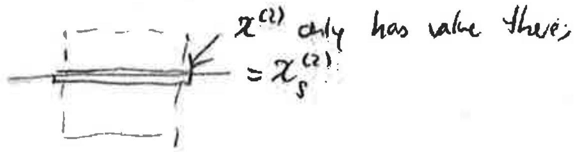
$$\frac{C\cdot m}{m^3} \quad \frac{C}{V\cdot m} \quad \frac{m}{V} \quad \frac{V}{m}\cdot\frac{V}{m}$$

<u>Surface</u> $P_s^{(2)} = \epsilon_0 \chi_s^{(2)} E^2$

$$\frac{C\cdot m}{m^2} \quad \frac{m^2}{V}$$

This unit for $\chi_s^{(2)}$ arise from the definition

$$\chi_s^{(2)} = \int \underset{m}{dz}\, \delta(z) \underset{\frac{m}{V}}{\chi^{(2)}}$$



7e. This will be the third-order susceptibility, mixing

$2\omega = \omega + \omega + 0$; $\chi^{(3)}_{ijk,z}(2\omega; \omega, \omega, 0)$ ← last index, non-resonant

$$P_i^{(3)}(2\omega, z) = \epsilon_0 \chi^{(3)}_{ijk,z}(2\omega; \omega,\omega,0)\, E_j(\omega) E_k(\omega) \cdot E_z(\omega, z)$$

$$P^{(3)}_{tot} = \int P_i^{(3)} dz = \epsilon_0 \chi^{(3)}_{ijk,z} \cdot E_j(\omega) \cdot E_k(\omega) \cdot \int_{z=0}^{\infty} E_z(0,z)\, dz \qquad \Phi_0 = -\int_{\infty}^{0} E_z(0)\, dz$$

$$= \epsilon_0 \chi^{(3)}_{ijk,z} \cdot E_j(\omega) \cdot E_k(\omega) \cdot \Phi_0$$

; assuming that $E_{j,k}$ do not change when E_z changes (otherwise $\int E_j E_k \frac{d\Phi}{dz} dz$)

Further symmetry restrictions apply for an isotropic material.
The number of tensor elements for χ_{ijkl} is here limited because l is only directed in the z direction, as there is only a electrostatic field along z. The number of ~~par~~ non-zero $\chi^{(3)}$ elements is not changed by it, just the number of possible polarization components. (E_{DC} is a participating field; not a material property). Ultimately it arises from the material but we don't care about the origin.

participating