7a

KDP

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

even numbers of (1,2) are necessary:

- 113, 223 + permutations
- 123, 132 + permutations
- 333

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

113 → −223

123 → 213

312 → 321

132 → 231

$$\chi_{113} = D_{21} D_{21} D_{33} \chi_{223}$$

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

- ~~113~~, ~~223~~ + ~~permutations~~
- 123, 132 + permutations
- ~~333~~

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

does not limit C_2 more than S_4 already did

123 → 213

⇒ final result for KDP

$$\boxed{\begin{aligned} xyz &= yxz \\ zxy &= zyx \\ xzy &= yzx \end{aligned}}$$

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

−1	+1	−1	(S_4)
+1	+1	+1	(σ_d)

7 b

Cubic crystal – for Q (432)

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

$D_{11} = D_{23} = 1$ $\quad D_{13} = -1$ $\quad D_{21} = -1$

$D_{32} = -1$ $\quad D_{22} = D_{31} = 1$ $\quad D_{12} = D_{33} = 1$

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

$C_{4,x}$

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
11	32	11	131 = −121	xyx = xzx = 0; $C_{4,y}$ $C_{4,z}$!

yxy = yzy = 0
zxz = zyz = 0

even 23 no effect

11	23	23	122 = 133
11	32	32	133 = 122
23	11	23	212 = 313

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

$C_{4,x}$

23	23	32	223 = −332
32	32	23	332 = 223
23	32	23	yzy = − zyz
32	23	32	232 = −323 ; 323 = 232 ; zyz = yzy] 0
23	32	32	233 = 322
32	23	23	322 = − 233

yyz = −zzy = 0
yzy = zyz = 0
yzz = zyy = −yzz = 0
all 0

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

11	23	32	123 = −132
11	32	23	132 = −123

] xyz and all permutations are ~~allowed~~ allowed

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 ~~the~~ the type xxx, x – x, – xx, xx – and permutations are zero.

$C_{4,z,y}$

xyz = − xzy , yxz = − zxy , yzx = − zyx with $C_{4,x}$. $C_{4,y}$, $C_{4,z}$? →

$D(C_4)_z$.

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

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		~~1~~ 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

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

$\chi^{(2)}$ only has value there; $= \chi_s^{(2)}$

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 participating tensor elements for χ_{ijkl} is here limited because l is only directed in the z direction, as there is only an electrostatic field along z. The number of ~~per~~ 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.