For any given symmetry species, this relation can be used to deduce the matrix form of the first-order field-induced Raman tensors from the tensors given in Table 2.3.3.1.

Example: We consider again the $4mm$ class crystal. The representation $\Gamma(\mathbf{f})$ of the magneto-optic tensor $\mathbf{f}$ in the $4mm$ class reduces as follows:

$$\Gamma(\mathbf{f}) = \Gamma_{\rm PV} \otimes \Gamma_{\rm PV} = 2{\rm A}_1 \oplus {\rm A}_2 \oplus {\rm B}_1 \oplus {\rm B}_2 \oplus 2{\rm E}.$$

Straightforward application of the mapping mentioned above then gives the following symmetry-restricted matrix forms of contributions to the magnetic-field-induced Raman tensors $\mathbf{R}^{jH}\mathbf{H}$ for all symmetry species of the $4mm$-class crystals. The number of independent parameters for each species is the same as in the intrinsic nonsymmetric zero-field Raman tensors:

$$A_1: \begin{pmatrix} . & ib'H_z & -ia'H_y \\ -ib'H_z & . & ia'H_x \\ ia'H_y & -ia'H_x & . \end{pmatrix}$$

$$A_2: \begin{pmatrix} . & . & ic'H_x \\ . & . & ic'H_y \\ -ic'H_x & -ic'H_y & . \end{pmatrix}$$

$$B_1: \begin{pmatrix} . & . & id'H_y \\ . & . & id'H_x \\ -id'H_y & -id'H_x & . \end{pmatrix}$$

$$B_2: \begin{pmatrix} . & . & -ie'H_x \\ . & . & ie'H_y \\ ie'H_x & -ieH_y & . \end{pmatrix}$$

$$E: \begin{pmatrix} . & ig'H_x & . \\ -ig'H_x & . & if'H_z \\ . & -if'H_z & . \end{pmatrix} \begin{pmatrix} . & ig'H_y & -if'H_z \\ -ig'H_y & . & . \\ if'H_z & . & . \end{pmatrix}.$$

Let us note that the conclusions mentioned above apply, strictly speaking, to non-magnetic crystals. In magnetic materials in the presence of spontaneous ordering (*ferro-* or *antiferromagnetic* crystals) the analysis has to be based on magnetic point groups.

2.3.4.4. *Stress- (strain-) induced Raman scattering*

Stress-induced Raman scattering is an example of the case when the external 'force' is a higher-rank tensor. In the case of stress, we deal with a symmetric second-rank tensor. Since symmetric stress ($\mathbf{T}$) and strain ($\mathbf{S}$) tensors have the same symmetry and are uniquely related *via* the fourth-rank *elastic stiffness tensor* ($\mathbf{c}$),

$$T_{\alpha\beta} = c_{\alpha\beta\mu\nu} S_{\mu\nu},$$

it is immaterial for symmetry purposes whether stress- or strain-induced effects are considered. The linear strain-induced contribution to the susceptibility can be written as

$$\Delta\chi_{\alpha\beta}(\mathbf{S}) = \left(\frac{\partial\chi_{\alpha\beta}}{\partial S_{\mu\nu}}\right) S_{\mu\nu}$$

so that the respective strain coefficients (conventional symmetric scattering) transform evidently as

$$[\Gamma_{\rm PV} \otimes \Gamma_{\rm PV}]_S \otimes [\Gamma_{\rm PV} \otimes \Gamma_{\rm PV}]_S,$$

i.e. they have the same symmetry as the *piezo-optic* or *elasto-optic* tensor. Reducing this representation into irreducible components $\Gamma(j)$, we obtain the symmetry-restricted form of the linear strain-induced Raman tensors. Evidently, their matrix form is the same as for quadratic electric-field-induced Raman tensors. In centrosymmetric crystals, strain-induced Raman scattering (in any order in the strain) is thus allowed for even-parity modes only.

2.3.5. Spatial-dispersion effects

For $\mathbf{q} = 0$, the normal modes correspond to a homogeneous phonon displacement pattern (all cells vibrate in phase). Phenomenologically, the $\mathbf{q}$-dependence of Raman tensors can be understood as a kind of morphic effect due to the gradients of the displacement field. Developing the contribution of the long-wavelength jth normal mode to the susceptibility in Cartesian components of the displacement of atoms in the primitive cell and their gradients, we obtain

$$\delta\chi^{(j)}_{\alpha\beta}(\mathbf{q}) = \sum_\kappa \left(\frac{\partial\chi_{\alpha\beta}}{\partial u^{(j)}_{\kappa,\gamma}}\right)_0 u^{(j)}_{\kappa,\gamma}(\mathbf{q}) + i\sum_\kappa \left(\frac{\partial\chi_{\alpha\beta}}{\partial(\nabla u^{(j)}_{\kappa,\gamma})_\delta}\right)_0 q_\delta u^{(j)}_{\kappa,\gamma}(\mathbf{q}), \quad (2.3.5.1)$$

where the derivatives are taken at $\mathbf{q} = 0$, and we use the obvious relation $\nabla u^{(j)}_{\kappa,\gamma} = i\mathbf{q}u^{(j)}_{\kappa,\gamma}$.

Transforming to normal coordinates, using (2.3.3.1), we identify the $\mathbf{q} = 0$ intrinsic Raman tensor $\mathbf{R}^{j0}$ of the jth normal mode, explicitly expressed *via* Cartesian displacements of atoms,

$$R^{jo}_{\alpha\beta} \equiv \chi^{(j)}_{\alpha\beta}(0) \equiv \left(\frac{\partial\chi_{\alpha\beta}}{\partial Q_j}\right) = \sum_\kappa \left(\frac{\partial\chi_{\alpha\beta}}{\partial u^{(j)}_{\kappa,\mu}}\right) \frac{e_{\kappa,\mu}(0,j)}{\sqrt{Nm_\kappa}}, \quad (2.3.5.2)$$

and introduce the first-order $\mathbf{q}$-*induced* atomic displacement Raman tensor coefficients $\mathbf{R}^{jq}$:

$$\begin{aligned} R^{jq}_{\alpha\beta\gamma} &\equiv -i\left(\frac{\partial\chi^{(j)}_{\alpha\beta}}{\partial q_\gamma}\right) = -i\left(\frac{\partial^2\chi_{\alpha\beta}}{\partial Q_j\partial q_\gamma}\right) \\ &= \sum_\kappa \left(\frac{\partial\chi^{(j)}_{\alpha\beta}}{\partial(\nabla u_{\kappa,\mu})_\gamma}\right)\frac{e_{\kappa,\mu}(0,j)}{\sqrt{Nm_\kappa}}. \end{aligned} \quad (2.3.5.3)$$

Hence, to the lowest order in $\mathbf{q}$, the transition susceptibility is expressed as

$$\delta\chi^{(j)}_{\alpha\beta}(\mathbf{q}) \cong \left(R^{j0}_{\alpha\beta} + iR^{jq}_{\alpha\beta\gamma}q_\gamma\right)Q_j(0). \quad (2.3.5.4)$$

In a more general case, spatial dispersion should be considered together with the electro-optic contributions due to the internal macroscopic field $\mathbf{E}$ and its gradients. Assuming the linear susceptibility to be modulated by the atomic displacements Q_j and the macroscopic electric field $\mathbf{E}$ as well as by their gradients ∇Q_j and $\nabla\mathbf{E}$, we can expand the transition susceptibility of the jth phonon mode $Q_j(\mathbf{q})$ to terms linear in $\mathbf{q}$ and formally separate the atomic displacement and electro-optic parts of the Raman tensor [see (2.3.3.15)]:

$$\begin{aligned} \delta\boldsymbol{\chi}^{(j)}(\mathbf{q}) &= (\partial\boldsymbol{\chi}/\partial Q_j)Q_j(\mathbf{q}) + i(\partial\boldsymbol{\chi}/\partial\nabla Q_j)\mathbf{q}Q_j(\mathbf{q}) \\ &\quad + (\partial\boldsymbol{\chi}/\partial\mathbf{E})\mathbf{E}^j(\mathbf{q}) + i(\partial\boldsymbol{\chi}/\partial\nabla\mathbf{E})\mathbf{q}\mathbf{E}^j(\mathbf{q}), \end{aligned}$$

or concisely

$$\delta\boldsymbol{\chi}^{(j)}(\mathbf{q}) = \mathbf{a}^j(\mathbf{q})Q_j(\mathbf{q}) + \mathbf{b}(\mathbf{q})\mathbf{E}^j(\mathbf{q}),$$

with

$$\mathbf{a}^j(\mathbf{q}) = (\mathbf{a}^{j0} + i\mathbf{a}^{jq}\mathbf{q}), \quad \mathbf{b}(\mathbf{q}) = (\mathbf{b}^0 + i\mathbf{b}^q\mathbf{q}). \quad (2.3.5.5)$$