Hidden Symmetry of Flexoelectric Coupling

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Abstract

Considering the importance of the flexoelectric coupling for the physical understanding of the gradient-driven couplings in mesoscale and nanoscale solids, one has to determine its full symmetry and numerical values. The totality of available experimental and theoretical information about the flexocoupling tensor symmetry (specifically the amount of measurable independent components) and numerical values is contradictory. However, the discrepancy between the theory and experiment can be eliminated by consideration all possible inner symmetries of the flexocoupling tensor and physical limits on it components values. Specifically, this study reveals the inner "hidden" symmetry of the static flexoelectric tensor that allows minimizing the number of its independent components. Revealed hidden symmetry leads to nontrivial physical sequences, namely it affects on the upper limits of the static flexocoupling constants. Also we analyze the dynamic flexoelectric coupling symmetry and established the upper limits for the numerical values of its components. These results can help to understand and quantify the fundamentals of the gradient-type couplings in different solids.

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I. INTRODUCTION

The flexoelectric coupling is principally important for fundamental insight into the complex electromechanics of meso-, nanoscale inorganic solids and biological systems, for which the strong strain gradients are inevitable present at the surfaces, interfaces, around point and topological defects [1, 2, 3, 4, 5, 6, 7].

Static flexoelectric coupling. The static flexoelectric effect is the appearance of elastic stress $\sigma_{ij}$ in response to electric polarization gradient $\partial P_i / \partial x_i$ (direct effect), and, vice versa, the polarization $P_i$ appears as a response to the strain gradient $\partial u_{ij} / \partial x_i$ (inverse effect) [8, 9, 10]. Variation of corresponding Lifshitz invariant included to the free energy functional of a solid [11],

$$F_L = \int d^3 r \frac{1}{2} \left( P_k \frac{\partial u_{ij}}{\partial x_k} - u_{ij} \frac{\partial P_k}{\partial x_k} \right)$$

leads to the linear relations,

$$\sigma_{ij} = f_{ijkl} \frac{\partial P_k}{\partial x_l}, \quad \quad P_k = f_{ijkl} \frac{\partial u_{ij}}{\partial x_l}.$$  (1b)

All components of the flexocoupling tensor $f_{ijkl}$ are symmetrical with respect to the first pair of indices, $f_{ijkl} = f_{jikl}$, and the trivial fact originates from the internal symmetry of the strain tensor, $u_{ij} = u_{ji}$, that in turn follows from its definition in continuum media approximation, $u_{ij} = (\partial U_i / \partial x_j + \partial U_j / \partial x_i) / 2$, via displacement vector components $U_i$. No other relations between its components follow from Eqs.(1).

Notably, the static flexoelectric effect is allowed by symmetry in all 32 crystalline point groups, because the strain gradient breaks the inversion symmetry, making the static flexoeffect omnipresent. For instance, Shu et al. [12], Quang and He [13], consider possible symmetries of the flexoelectric tensor and derived the number of its independent components for each symmetry. Let us underline the fact that Shu et al., Quang and He used the invariance of the flexocoupling tensor $f_{ijkl}$ to the permutation of the first one pair of indices, $f_{ijkl} = f_{jikl}$. This work is about another nontrivial fact, that $f_{ijkl} = f_{iklj}$ (the derivation is given below in Section II). Hence both the trivial ($f_{ijkl} = f_{jikl}$) and nontrivial ($f_{ijkl} = f_{iklj}$) equalities are valid.

It has been shown that the static flexocoupling induces imprint [14, 15, 16], internal bias [17, 18] and dead layer effect [19, 20] in ferroelectric thin films, and vortices in superlattices [21]. Flexoelectric coupling strongly changes the structure and electro-transport properties of the domain walls and interfaces in ferroelectrics [22, 23, 24, 25] and ferroelastics [26, 27, 28]. The flexocoupling leads to the hardening of solids at nano-indentation [29, 30, 31], significantly affects on the local electrochemical strains appeared in materials with mobile charges [32, 33], as well as on the...
mechanical writing of ferroelectric polarization by the tip [34]. The flexocoupling can induce incommensurate spatially modulated phases in many ferroics including antiferroelectric and antiferrodistortive ones [28, 35, 36, 37]. Since a wave excitation of any nature is impossible without a local gradient of the corresponding physical quantity [38, 39], the flexoelectric effects significantly influence the propagation of surface acoustic waves in non-piezoelectric solids [40].

Considering the importance of the flexoelectricity for the physical understanding of the gradient-driven mesoscale and nanoscale couplings in solids, one has to determine the internal symmetry and numerical values of static flexocoupling tensor $f_{ijkl}$. Numerical values of the static flexocoupling tensor. Typically the values $f_{ijkl}$ calculated from the first principles [41, 42, 43, 44, 45] can be several orders of magnitude smaller than those measured experimentally [46, 47, 48, 49]. The discrepancy motivated Yudin, Ahluwalia and Tagantsev [50] to establish theoretically the upper limits for the values of the static flexoelectric coefficients $f_{ijkl}$ in solids with cubic parent phase symmetry. The calculated maximal values of $f_{ijkl}$ showed that the anomalously high flexoelectric coefficients measured for perovskite ceramics [46, 47, 48] cannot be related with the manifestation of the static flexoelectric effect. Morozovska et al. [51, 52, 53] established that spatially modulated phases appear and become stable in commensurate ferroelectrics if the flexocoupling constants exceed the maximal critical values, which depend on the electrostriction and elastic constants, temperature, and gradient coefficients in the Landau-Ginzburg-Devonshire functional. Hence the comparison of the aforementioned experimental and theoretical results tell us that numerical values of the static flexocoupling tensor are rather contradictory, indicating on a limited understanding of the effect strength.

II. HIDDEN SYMMETRY OF THE FLEXOCOUPLING TENSOR

Specifically, for one of the highest m3m cubic point symmetry group three independent components of the flexocoupling tensor are $f_{1111} = f_{2222} = f_{3333}, f_{1122} = f_{1133} = f_{2233} = f_{3322} = \ldots$ and the third component $f_{1212} = f_{1313} = f_{2323} = f_{2121} = \ldots$ that is identically equal to $f_{1221} = f_{1331} = f_{2313} = f_{2112} = \ldots$ due to the permutation symmetry, $f_{ijkl} \equiv f_{jikl}$.

At first Zubko et al. [49] measured experimentally the tensor of flexoelectric effect in SrTiO$_3$ with cubic parent phase m3m symmetry and stated that it includes all three components, $f_{1111}, f_{1122}$ and $f_{1212}$. The flexoelectric coefficients measurement [49] is the dynamical bending of thin electrode plates using “three knives” setup with simultaneous measurement of a displacement current. The applied stain has zero average value and changes its sign inside the plate (i.e. distribution with “pure
The induced polarization is proportional to the strain gradient value determined from the geometry of deformed system.

However, later Zubko et al. [54] recognized that it was mathematically impossible to define all three components of the flexocoupling tensor from the quasi-static bending of plates [49] alone and suggested to use the results of independent dynamical measurements based on Brillouin scattering by Hehlen et al. [55] (see also the work of Tagantsev et al. [56]). Specifically, Zubko et al. wrote in the erratum [54] to their earlier paper [49] that "despite the use of different sample geometries and crystallographic orientations, only two independent equations involving \( f_{1111}, f_{1122}, \) and \( f_{1212} \) can be obtained from bending experiments alone, and additional information is required to find the individual tensor components.” At the same time additional equation for \( f_{1212} \) was obtained from independently measured value based on Brillouin scattering data [55]. It should be noted that obtained in this way values of \( f_{1122} \) and \( f_{1212} \) appeared close to each other (7 nC/m and 5.8 nC/m respectively), especially in comparison with \( f_{1111} = 0.2 \) nC/m.

It should be also noted that Yudin et al. [50] found only two independent conditions for upper limits for three independent flexoelectric tensor components \( f_{1111}, f_{1122}, \) and \( f_{1212} \) required for the homogeneous phase stability under the absence of higher elastic gradients. From the phenomenological theory of phonon dispersion relations (see e.g. [51]) not all the components of static flexocoupling tensor are involved. Hong and Vanderbilt [43] calculated flexoelectric coefficients under the assumption that “transversal” flexocoupling coefficient, \( f_T = f_{1122} - f_{1212} \) is zero. Below we will prove the assumption \( f_{1122} - f_{1212} \equiv 0 \) by exploring the "hidden" symmetry of the flexocoupling.

Hence, the primary goal of this work is to establish the inner "hidden" symmetry of the static flexocoupling tensor that allows minimizing the number of its independent components. Actually, the integration in parts for in Eq.(1a) for a bulk infinite solid yields:

\[
\int_{v \to \infty} d^3 r f_{ijkl} \frac{1}{2} \left( u_y \frac{\partial P_k}{\partial x_i} - P_k \frac{\partial u_y}{\partial x_i} \right) = \int_{v \to \infty} d^3 r f_{ijkl} u_y \frac{\partial P_k}{\partial x_i} = ... = \int_{i \to \infty} d^3 r \left( f_{ijkl} + f_{jikl} \right) \frac{\partial U_i}{\partial x_j} \frac{\partial P_k}{\partial x_l} + ... = \int_{i \to \infty} d^3 r \left( f_{ijkl} + f_{jikl} \right) \frac{\partial U_i}{\partial x_j} \frac{\partial P_k}{\partial x_l}.
\]

Detailed derivation of each step in Eq.(2) is given in Appendix A, where we dropped all surface integrals (e.g. \(- \int_S d^2 r \frac{f_{ijkl}}{2} u_y P_i n_j \)), regarding them negligible for an infinite bulk material in a continuum media approximation. This is correct, because the flexoelectric tensor \( f_{ijkl} \) describes the macroscopic bulk properties of the material in a continuum media approximation and therefore its symmetry cannot depend on the surface surrounding the sample.
Since the summation in Eq.(2) is performed on the "dumb" indices, they could be redefined in arbitrary manner (see Appendix A), so that one concludes that the latter equality in Eq.(2) is possible for the arbitrary polarization and displacement only if

\[ f_{ijkl} = f_{iklj} \cdot \]  \hspace{1cm} (3)

Nontrivial relation Eq.(3) along with the trivial relation \( f_{ijkl} = f_{jikl} \) lead to the conclusion that only two components \( f_{1111} \) and \( f_{1122} = f_{1221} = f_{1122} \) are independent for m\( m \) symmetry (instead of three or four components as regarded previously).

Note, that for the isotropic media only one non-trivial component of tensor \( f_{ijkl} \) is possible in contrast to findings of Le Quang and He [13], who did consider additional internal symmetry of flexoelectric tensor.

That say Eq.(3) is a manifestation of the static flexoeffect "hidden" symmetry. The conclusion can explain previously unexplained conclusion from Zubko et al experiments [49] and Hong and Vanderbilt ab initio calculations [43] for materials with cubic m\( m \) symmetry. Specifically for m\( m \) the effective flexoelectric coefficients \( f'_{1111}(\phi) = f_{1111} - \mu \sin^2(2\phi) \) and \( f'_{1212}(\phi) = f_{1212} + \mu \sin^2(2\phi) \) can be measured experimentally as a function of sample rotation angle \( \phi \). The anisotropy value \( \mu = (f'_{1111} - f_{1122} - 3f_{1212})/2 \) is equal to \( (f_{1111} - 3f_{1212})/2 \) because \( f_{1122} = f_{1212} \) accordingly to Eq.(3).

From these relations the "true" flexocoefficients are only \( f'_{1111} = f'_{1111}(0) \) and \( f'_{1212} = f'_{1212}(0) \). Since \( f'_{1212}(\pi/4) = (f_{1111} - f_{1212})/2 \) we obtain that \( 2f'_{1212}(\pi/4) \equiv f'_{1111}(0) - f'_{1212}(0) \) as the direct consequence of Eq.(3), and the latter relation can be verified experimentally. The angular dependences \( f'_{1111}(\phi) \) and \( f'_{1212}(\phi) \) are shown in Fig. 1.

Since relation (3) is based on the transformation properties of the phenomenological free energy of the bulk material in a continuum media approximation, it has nothing similar with e.g. (semi)-microscopic theories [57], predicting Cauchy relations, \( c_{ijkl} = c_{jikl} \), for elastic stiffness \( c_{ijkl} \) [58].
For symmetries lower than cubic the revealed "hidden" symmetry described by Eq.(3) leads to nontrivial physical sequences, namely it should affect on the upper limits of the static flexocoupling constants established by Yudin et al [50]. To illustrate the statement let us analyze the expression derived in Ref.[33] for the Fourier spectra of linear static dielectric susceptibility $\chi_{ij}(r)$. The expression for inverse susceptibility, $\frac{1}{\chi_{ij}}(k)$, valid for a dielectric solid (or in a paraelectric phase of ferroelectric), has a relatively simple form:

$$\frac{1}{\chi_{ij}}(k) = 2\alpha \delta_{ij} + (g_{ij} - f_{\text{min}} - f_{\text{eff}} k_i k_j S_{ij}(k)) k_i k_j .$$

The summation in Eq.(4) is performed over all repeating indexes allowing for Eq.(3). The coefficient $\alpha$ is positive and temperature dependent in the paraelectric phase of ferroelectrics as $\alpha = \alpha_T (T - T_C)$. $g_{ijkl}$ is a positively defined symmetrized tensor of polarization gradient coefficients [33]. Inverse matrix is $S^{-1}_{ik}(k) = c_{ijk} k_i k_j$ and elastic compliances permutative symmetry is $c_{ijkl} = c_{ikjl} = c_{ijlk}$. The equations $[g_{ij} - f_{\text{min}} - f_{\text{eff}} k_i k_j S_{ij}(k)] k_i k_j = 0$ ($i, j = 1, 2, 3$) define zeroes $\frac{1}{\chi_{ij}}(k) = 0$ at $\alpha = 0$. For the wave vector $k = (0, 0, k)$ directed along direction $[0, 0, 1]$ the matrix $S^{-1}_{ik}(k) = c_{ijk} k^2$ and so $S_{ij}(k) = S^{(3)}_{ij}/k^2$ at that $c_{ijk} S^{(k)}_{ij} = S_{ij}$. In this way the stability conditions for $[1, 0, 0]$, $[0, 1, 0]$ and $[0, 0, 1]$ directions of the wave vector acquire the form

$$f_{mkk} f_{ijk} S^{(k)}_{mkl} \leq g_{ijk} \quad \text{(without summation over } k = 1, 2, 3).$$
and define the upper limits of the static flexocoupling constants required for the homogeneous state stability for arbitrary point symmetry.

The explicit form of the stability conditions for the flexoelectric coefficients depends on the direction of the wave vector of the fluctuations and concrete symmetry. In order to show the role of the flexocoupling hidden symmetry we derived the stability conditions imposed on $f_{ijkl}$ values for m3m symmetry, transverse (TO) and longitudinal (LO) optic modes propagation in [100], [110] and [111] directions. Results are listed them in the second column of Table I. For m3m symmetry Yudin et al [50] considered [100], [110] and [111] k-directions for TO mode (see the the third column of Table I).

For the direction [100] and [110], our conditions for TO mode coincide with the conditions obtained by Yudin et al. For the direction [111] our condition has more simple form than the one derived by Yudin et al, since the additional constraint, $f_{1212} \equiv f_{1122}$, is valid accordingly to Eq.(3).

| Wave vector direction | This work gives the following relations for m3m cubic symmetry in matrix notations | Yudin et al. [50] obtained relations for m3m cubic symmetry in matrix notations |
|-----------------------|-------------------------------------------------|--------------------------------------------------|
| [100]                 | $\mathbf{TO-mode:} \quad f_{1212}^2 \leq c_{1212}g_{1212}$ | $\mathbf{TO-mode:} \quad f_{1212}^2 \leq c_{1212}g_{1212}$ |
|                       | $\mathbf{LO-mode:} \quad f_{1111}^2 \leq c_{1111}g_{1111}$ | LO-mode was not considered |
| [110]                 | $\mathbf{TO-mode:} \quad (f_{1111} - f_{1122})^2 \leq (c_{1111} - c_{1122})(g_{1111} - g_{1122})$ | $\mathbf{TO-mode} \quad (f_{1111} - f_{1122})^2 \leq (c_{1111} - c_{1122})(g_{1111} - g_{1122})$ |
|                       | $\mathbf{LO-mode} \quad (f_{1111} + 3f_{1212})^2 \leq (c_{1111} + c_{1122} + 2c_{1212})(g_{1111} + g_{1122} + 2g_{1212}) \leq$ | LO-mode was not considered |
| [111]                 | $\mathbf{TO-mode} \quad f_{1111}^2 \leq (c_{1111} - c_{1122} + c_{1212})(g_{1111} - g_{1122} + g_{1212})$ | $\mathbf{TO-mode} \quad (f_{1111} - f_{1122} + f_{1212})^2 \leq (c_{1111} - c_{1122} + c_{1212})(g_{1111} - g_{1122} + g_{1212})$ |
|                       | $\mathbf{LO-mode} \quad (f_{1111} + 6f_{1212})^2 \leq (c_{1111} + 2c_{1122} + 4c_{1212})(g_{1111} + 2g_{1122} + 4g_{1212}) \leq$ | LO-mode was not considered |

### III. INNER SYMMETRY AND UPPER LIMITS OF DYNAMIC FLEXOELECTRIC EFFECT

From considerations of the symmetry theory stating that all terms and invariants, which existence does not violate the symmetry of the system, are allowed, Tagantsev et al. (see reviews [2, 3] and refs therein) predicted the existence of dynamic flexoelectric effect originated from the cross-term in the kinetic energy, proportional to polarization components $P_i$ and elastic displacement $U_j$ time derivatives,
\[
\chi_{ij} (t, \mathbf{r}) = \alpha - \mu \omega^2 \delta_{ij} + g_{ijkl} k^i k^l - \left( f_{\text{min}} k^i k^l - M_m^2 \omega^2 \right) \left( f_{\text{max}} k_i k_j - M_p^2 \omega^2 \right) S_{\text{mi}} (\mathbf{k}, \omega)
\]

(7)

The summation in Eq.(7) is performed over all repeating indexes. Inverse matrix
\[
S_{\text{ij}}^{-1} (\mathbf{k}, \omega) = c_{ijkl} k_i k_j - \rho \omega^2 \delta_{ij} \quad (\delta_{ij} \text{ is the Kronecker symbol}), \rho \text{ is the density of a solid and } \mu \text{ is a kinetic coefficient. The coefficient } \alpha \text{ is positive for dielectrics and is temperature dependent in the paraelectric phase of ferroelectrics (e.g. } \alpha = \alpha_T (T - T_C) \text{ in the vicinity of Curie temperature } T_C), g_{ijkl} \text{ is a positively defined symmetrized tensor of polarization gradient coefficients [33]. The solutions of characteristic equation } \tilde{\chi}_{ij}^{-1} (\mathbf{k}, \omega) = 0 \text{ give the optic and acoustic soft phonon dispersion law in the parent phase of ferroics. Putting condition } \mathbf{k} = 0 \text{ in Eq.(7) we obtain the equations}
$2\alpha\delta_{ij} + \left(\frac{M_{jj'}M_{jj'}}{\rho} - \mu\delta_{ij}\right) \omega^2 = 0$. The nontrivial solutions of these equations for $\alpha \geq 0$ correspond to the values of optic mode at $k = 0$,

$$\omega^{opt}_i(k = 0) = \frac{2\alpha(T)}{\mu - (M_{1i}^2 + M_{2i}^2 + M_{3i}^2)/\rho}, \quad (i=1, 2, 3). \quad (8)$$

For diagonal tensor $M_{ij} = M_{ii} \delta_{ij}$, the solution (8) exists under the conditions

$$M_{ii}^2 < \mu\rho \quad \text{(without summation over } i=1, 2, 3). \quad (9)$$

For cubic m3m symmetry the dynamic flexocoupling tensor is diagonal, $M_{11} = M_{22} = M_{33} = M$, and the inequality $M^2 < \mu\rho$ follows from Eq.(9). Numerical estimates give $|M_{11}| < 11.3 \times 10^{-8} \text{ V s}^2/\text{m}^2$ for $\mu = 1.59 \times 10^{-18} \text{ s}^2\text{Jm}$, $\rho = 7.986 \times 10^3 \text{ kg/m}^3$ (corresponding to PbTiO$_3$ in a cubic paraelectric phase at $T = 783 \text{ K}$, $T_C = 752 \text{ K}$); and $|M_{11}| < 32.9 \times 10^{-8} \text{ V s}^2/\text{m}^2$ for $\mu = 22 \times 10^{-18} \text{ s}^2\text{Jm}$, $\rho = 4.930 \times 10^3 \text{ kg/m}^3$ (corresponding to paraelectric SrTiO$_3$ at $T = 120 \text{ K}$). The values of $|M_{11}|$ corresponding to the best fitting of experimentally measured soft phonon spectra appeared significantly lower, namely $|M_{11}| = 2 \times 10^{-8} \text{ V s}^2/\text{m}^2$ for PbTiO$_3$ and $|M_{11}| = 22 \times 10^{-8} \text{ V s}^2/\text{m}^2$ for SrTiO$_3$ [33].

The temperature dependence of expression (8) can serve for determination of dynamic flexocoupling value [compare curves 1-4 in Fig. 2(a)], because any deviation of the ratio $\eta = \mu(\omega^{opt})^2/2\alpha(T - T_C)$ from unity at $T > T_C$ can follow from the existence of the temperature-dependent term in denominator, $(M_{ii}^2 + M_{2i}^2 + M_{3i}^2)/\rho$. Almost horizontal solid lines 2, 3 and 4 in Fig. 2(b) correspond to $\eta$ values calculated for different $M_{ij} \neq 0$. They essentially differ from $\eta = 1$ (dashed line 1) calculated for $M_{ij} = 0$. Also we regard that $\mu$ and $M_{ii}$ are temperature independent, $\alpha = \alpha_T(T - T_C)$ and the density $\rho = \rho_T[1 + \beta_T(T_C - T)]$ obeys linear thermal expansion law; $\beta_T$ is the thermal expansion coefficient.
FIG. 2. Temperature dependence of the soft phonon TO frequency $\omega^{\text{opt}}$ (a) and the ratio (b) $\eta = \mu (\omega^{\text{opt}})^2 / 2\alpha_T (T - T_c)$ calculated from Eq.(8) at $M_{ij} = 0$ (dashed black curve) and $|M_{ij}| = (2, 5, 8) \times 10^{-8}$ V s$^2$/m$^2$ (solid red, magenta and blue curves), $\alpha_T = 3.765 \times 10^5$ F/m, $T_c = 752$ K, $\mu = 1.59 \times 10^{-18}$ s$^2$Jm, $\rho = 7.986 \times 10^3$ kg/m$^3$ and $\beta_T = 3.2 \times 10^{-5}$ K$^{-1}$ corresponding to PbTiO$_3$ in a paraelectric phase.

IV. CONCLUSION

To resume, this study revealed the inner "hidden" symmetry of the static flexoelectric tensor that allows minimizing the number of its independent components. Revealed hidden symmetry leads to nontrivial physical sequences, namely it affects on the upper limits of the static flexocoupling constants. Also we analyze the dynamic flexoelectric coupling symmetry and established the upper limits for the numerical values of its components. These results can help to understand and quantify the fundamentals of the gradient-type flexocoupling.

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APPENDIX A. Derivation of relation (3)

Let us consider the contribution of flexoelectric coupling to the free energy

$$\int d^3 r \left\{ \frac{1}{2} \left( \frac{\partial P_k}{\partial x_i} - P_k \frac{\partial u_{ij}}{\partial x_i} \right) u_{ij} \right\} = \int d^3 r \int_{-\infty}^{\infty} \frac{f_{ijkl} \partial P_k}{\partial x_i} \frac{\partial u_{ij}}{\partial x_i} \left( \frac{\partial P_k}{\partial x_i} \right) \right\} = \int d^3 r f_{ijkl} u_{ij} \frac{\partial P_k}{\partial x_i} \right\}$$

(A.1)
When integrating in parts in Eq.(A.1) we neglected the surface integrals like \[ \int d^2r \frac{f_{ijkl}}{2} u_{ij} P_k n_l \]
hereinafter, since it can be done for a bulk material in a continuum media approximation. Since 
\[ u_{ij} = \left( \partial U_i / \partial x_j + \partial U_j / \partial x_i \right) / 2 \], the transformations of Eq.(A.1) yields 
\[ \int d^3r \frac{f_{ijkl}}{2} \left( \frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i} \right) \frac{\partial P_k}{\partial x_l} = \int d^3r \left( f_{ijkl} + f_{jikl} \right) \frac{\partial U_i}{\partial x_j} \frac{\partial P_k}{\partial x_l} \]

Comparing underlined by "blue" and "red" steps of the transformations (A.2) for an arbitrary coordinate-dependent function 
\[ \frac{\partial U_j}{\partial x_i} \frac{\partial P_k}{\partial x_l} \], one leads to the relation 
\[ f_{ijkl} + f_{jikl} = f_{inkj} + f_{inkj} \], \hspace{1cm} (A.3) 
since \[ f_{ijkl} = f_{jikl} \] and \[ f_{inkj} = f_{inkj} \], due to the symmetry of the strain tensor, \[ u_{ij} = u_{ji} \], Eq.(A.3) elementary leads to Eq.(3):
\[ f_{ijkl} = f_{jikl} = f_{inkj} = f_{inkj} \] \hspace{1cm} (A.4)

Hence we proved that besides the relation \[ f_{ijkl} = f_{jikl} \] there is one more relation \[ f_{ijkl} = f_{inkj} \], Eq. (3), that imposes additional constrains on the structure of the flexoelectric tensor.

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