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SIMULATION OF LONG-WAVELENGTH OPTICAL PHONONS BY GENERALIZED INTERNAL STRAINS

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Abstract. A generalized external force applied to a crystal can introduce new atomic position parameters known as internal strains. Each internal strain may be looked as a "frozen" \( q = 0 \) optical phonon of appropriate symmetry. Explicit forms can be worked out for the internal strains produced by any type of force and crystal class. Related crystal parameters can be determined in any case with X-ray diffraction techniques.

The concept of internal strain will be generalized to include all possibilities of microscopic relative displacements of the various Bravais sublattices which are produced by any type of external force \( F \). When \( F \) is applied to the crystal a macroscopic strain \( \eta \) may be produced. The mechanism which produces \( \eta \) depends on the nature of \( F \) [e.g. piezoelectricity (electrostriction) when \( F \) is an electric field \( E \) in first (second) order, piezomagnetism (magnetostriction) when \( F \) is a magnetic field \( H \) in first (second) order, etc]. Because of \( F \) the \( k \)th atom in the \( i \)th cell moves from its initial position \( r_{i,k} \) to a new one

\[
r_{i,k} \approx (1 + \eta) r_{i,k} + u_k
\]

The first term is the macroscopic contribution to \( r_{i,k} \) while the second term \( (u_k) \) represents a microscopic strain due to \( F \). It is the same for all atoms of type \( k \) in the crystal, i.e., it represents a microscopic displacement of the entire \( k \)th Bravais sublattice and as such it constitutes an internal strain. Both terms in (1) depend on the symmetry of the crystal independently of each other.

There are \( 3(N-1) \) different sublattice displacements, where \( N \) the no. of atoms per unit cell. \( 3(N-1) \) also gives the no. of \( q = 0 \) optical phonons of the crystal. The normal coordinate \( u_j \) of the phonon \( j \) and the internal strain \( u_k \) (or \( u_{k,\lambda} \), \( \lambda=x,y,z \)) are qualitatively similar in the sense that both represent sublattice displacements and transform alike under the crystal symmetry operations. When "frozen" each phonon will look exactly like one of the \( u_k \)'s. The symmetry of \( u_j \) will determine the type of \( F \) which is necessary to produce its internal strain \( u_k \). This symmetry is described by one of the point group irreducible representations \( \Gamma(j) \). Thus \( \Gamma(j) = \Gamma(u_k) \). The various \( \Gamma(j) \)'s are obtain-

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ned from the reduction of $\Gamma_{opt}$, the reducible rep of the most general
displacement of the $q = 0$ optical phonon of the crystal.

The connection between $F$ and $u_k$ is derived thermodynamically. Let $f$ be the intensive parameter which corresponds to $F$. ($F$ is treated
as an extensive parameter\textsuperscript{2}). To first order of $u_k$ we can write

$$F = G \cdot u_k \quad \text{or} \quad f = g \cdot u_k \quad \text{(2)}$$

This results in a contribution to the internal energy of the crystal

$$\delta U(k) = F \cdot f = F \cdot g \cdot u_k = g_{\mu,\lambda} \cdot F_{\mu} \cdot u_{k,\lambda} \quad \text{(3)}$$

where $u_k$ may be a composite index. Since $\delta U(k)$ is a scalar, the only
products $F_{\mu} \cdot u_{k,\lambda}$ entering (3) are those which transform like scalars.
This allows one to determine the type $F$ necessary to produce a speci-
fic $u_k$ and vice versa. The criterion is that $\Gamma(j)$, or $\Gamma(u_k,j)$, be in-
cluded in the reduction of $\Gamma(F)$ at least once, in general $n_j$ times. The
number $n_j$ indicates the number of independent components of the
tensor coefficient $g$. The form of $g_{\mu,\lambda}$ is identical to that of known mo-
de coefficients\textsuperscript{3}. Typical examples for various forces are given next.

1. Electric field $E$. The reduction $\Gamma(E)$ includes only IR-active pho-
nons. Eq. (3) yields $g_{\mu,\lambda} = (\partial^2 U(k) / \partial \mu \partial \lambda)_{\mu,\lambda}$, i.e., $g_{\mu,\lambda}$ transforms like the phonon effective charge tensor\textsuperscript{3} $e_{\mu,\lambda} = (\partial^2 U(j) / \partial \mu \partial \lambda)_{\mu,\lambda}$.

2. Elastic stress or strain $\eta$. The reduction $\Gamma(\eta)$ includes only Raman-
active phonons in symmetric scattering\textsuperscript{3,4}. Since $g_{\rho,\sigma,\lambda} = (\partial^2 U(k) / \partial \rho \partial \sigma \partial \lambda)_{\rho,\sigma,\lambda}$ 
it transforms like the Raman tensor $a_{j,\lambda} = (\partial^2 U(j) / \partial \rho \partial \lambda)_{j,\lambda}$.

3. Electric field gradient $\nabla \rho E_{\sigma} = (\partial E_{\rho} / \partial \sigma)_{\rho,\sigma}$. The reduction $\Gamma(VE)$ in-
cludes only Raman-active phonons in non-symmetric scattering. $g_{\sigma,\rho,\lambda}$ transforms like the non-symmetric Raman tensor\textsuperscript{3}.

4. Strain gradient $\nabla \rho \eta_{\sigma \tau}$. The reduction of $\Gamma(\nabla \eta)$ is the same as that of the piezo-electric tensor. $g_{\sigma \tau \rho,\lambda}$ transforms like the electric field-induced Raman tensor\textsuperscript{3}.

5. Hydrostatic pressure $P$. The reduction $\Gamma(P)$ coincides with $A_1$, the
totally symmetric rep. Hence there are as many internal strains $u_k$ pro-
duced by $P$ as the number of phonons $A_1$. This also is the same as the
number of atomic position parameters which cannot be uniquely deter-
mined by symmetry arguments\textsuperscript{4}. The tensor $g_{\mu,\lambda}$ becomes a scalar.

6. Magnetic field $H$. $u_{k,\mu,\lambda}$ will transform like an axial vector. $g_{\mu,\lambda}$ is
like a second-rank polar tensor which reverses sign upon time revers-
ral (c-type tensor\textsuperscript{5}). I can only exist for the 90 magnetic crystal
classes. The matrices of such tensors are given in tables 7 and 4 of
Ref.5.

As an example we consider $\alpha$-quartz (N=9 point group $D_3$). We have\textsuperscript{3}

$$\Gamma_{opt} = 4A_1 + 4A_2 + 8E \quad \Gamma(VE) = 2A_1 + A_2 + 3E \quad \Gamma(P) = A_1$$
$$\Gamma(E) = A_2 + E \quad \Gamma(H) = A_2 + E$$
$$\Gamma(\eta) = 2A_1 + 2E \quad \Gamma(\nabla \eta) = 2A_1 + 4A_2 + 6E$$
According to these reductions there are 16 independent internal strains grouped into 3 types, 4 of type \( A_1 \) (non-degenerate), 4 of type \( A_2 \) (non-degenerate) and 8 of type \( E \) (doubly degenerate). Type \( A_1 \) can be produced by \( \eta, V, E, P \) and \( \nabla \eta \) with 2,2,1,2 independent components each, for each of the corresponding tensors \( g \). Type \( A_2 \) can be produced by \( E, V, E \) and \( \nabla \eta \) with 1,1,4 components each for the tensors \( g \), etc. For a magnetic crystal with \( N=9 \) and magnetic structure \( D_3 \), a field \( H \) will produce 4 internal strains of type \( A_2 \) and 8 of type \( E \) with only one component each for the tensor \( g \). Once the forms of \( g \) are written, similar ones hold for \( G \) of (2) and its inverse \( Y \), where \( u = y \cdot F \). This is because \( g, G \) and \( Y \) have the same symmetry. We will consider the case of \( F = \nabla \eta \) for the \( A_2 \) type of internal strains of \( \alpha \)-quartz. \( Y \) follows from the electric field-induced Raman tensor, according to the scheme

\[
\sigma^{E}_{\sigma \tau \rho, \lambda} = (\varepsilon^\lambda u^{(j)}/\partial \epsilon_{\sigma}, \partial \sigma, \partial \rho, \partial \tau, \partial \lambda) \Rightarrow (\varepsilon^\lambda u^{(k)}/\partial \rho, \partial \tau, \partial \lambda) \Rightarrow y^{\sigma \tau \rho, \lambda}
\]

The results for \( Y \) and \( u = y \cdot \nabla \eta \) are

\[
\gamma^{\sigma \tau \rho, z} = \begin{pmatrix}
\gamma_1 & \ldots & \gamma_3 & 2 \gamma_2 \\
\gamma_2 & -\gamma_2 & \gamma_3 & \gamma_4 \\
\gamma_1 & \gamma_1 & \ldots & \gamma_4 \\
\end{pmatrix}
\]

\[
u^{A_2, z} = \gamma_1 \left( \frac{\partial \eta_{xy}}{\partial z} + \frac{\partial \eta_{yy}}{\partial z} \right) + \gamma_2 \left( \frac{\partial \eta_{xx}}{\partial y} - \frac{\partial \eta_{yy}}{\partial y} + 2 \frac{\partial \eta_{xy}}{\partial x} \right)
\]

\[
+ \gamma_3 \left( \frac{\partial \eta_{xx}}{\partial y} + \frac{\partial \eta_{xy}}{\partial y} \right) + \gamma_4 \left( \frac{\partial \eta_{xy}}{\partial y} \right)
\]

There are 4 sets of values for \( \gamma_1 \ldots \gamma_4 \) since we expect 4 independent internal strains of type \( A_2 \) due to \( \nabla \eta \). From (6) one can easily derive the selection rules for inducing a desired term of \( u^{A_2, z} \).

Internal strains due to external stresses have been studied for some materials through the observation of forbidden X-ray diffractions. In principle similar techniques can be employed to manifest internal strains due to forces other than stresses.

References