Relation between Interband Dipole and Momentum Matrix Elements

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<table>
<thead>
<tr>
<th>Infinite Volume Non-Vanishing Boundary Conditions</th>
<th>Finite Volume Periodic Boundary Conditions</th>
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<tbody>
<tr>
<td>$\langle c k'</td>
<td>r</td>
</tr>
<tr>
<td>Blount 1962</td>
<td>Incorrectly proven in Haug 1972</td>
</tr>
<tr>
<td>$\langle u_{c k}</td>
<td>\hat{p}</td>
</tr>
<tr>
<td>Adams 1952, Haug 1972</td>
<td>Haug 1972</td>
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<tr>
<td>$\langle u_{c k}</td>
<td>\hat{p}</td>
</tr>
<tr>
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<td>\hat{p}</td>
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<tr>
<td>Follows from above, see Gu et al. 2013</td>
<td>This work</td>
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</table>
In crystals with periodic boundary conditions:

\[
\hat{\mathbf{p}} = \frac{im}{\hbar} [H_0, \mathbf{r}]
\]

(classically: \( p = m\mathbf{v} \) )

\[
H_0 = -\frac{\hbar^2}{2m} \nabla^2 + V_L(\mathbf{r})
\]

\[
\langle c \mathbf{k}' | \hat{\mathbf{p}} | v \mathbf{k} \rangle_{\text{vol}} = \frac{im}{\hbar} \langle c \mathbf{k}' | H_0 \mathbf{r} - H_0 | v \mathbf{k} \rangle_{\text{vol}}
\]

\[
\langle c \mathbf{k}' | H_0 \mathbf{r} | v \mathbf{k} \rangle_{\text{vol}} = \mathcal{E}_{v \mathbf{k}} \langle c \mathbf{k}' | \mathbf{r} | v \mathbf{k} \rangle_{\text{vol}}
\]

\[
\langle c \mathbf{k}' | H_0 \mathbf{r} | v \mathbf{k} \rangle_{\text{vol}} \neq \mathcal{E}_{c \mathbf{k}} \langle c \mathbf{k}' | \mathbf{r} | v \mathbf{k} \rangle_{\text{vol}}
\]

Probably widely known, but not widely applied in context of periodic boundary conditions

Comparison: infinite system, non-vanishing boundary conditions

\[
\langle c \mathbf{k}' | \hat{\mathbf{p}} | v \mathbf{k} \rangle_{\infty} = im \omega_{c \mathbf{k}', v \mathbf{k}} \langle c \mathbf{k}' | \mathbf{r} | v \mathbf{k} \rangle_{\infty}
\]

- Valid in distribution sense
- Formal dipole matrix element is really that of the k-gradient operator
- k-gradient may not exist in case of degeneracy, see Zak 1985, Foreman 2000
- Diagonal element \( k = k' \) not defined
- Proof using limiting procedure with spatially limited wave-packets in Gu et al. 2013
Bulk GaAs: simple Cohen-Bergstresser pseudopotential approach

- Ga
- As

Pseudopotential:

s-like c-band wave fct.

\[ \begin{array}{c}
0.875 \\
0.375 \\
0.875 \\
0.375 \\
1.875 \\
-0.125 \\
0.375 \\
0.875 \\
1.875 \\
1.375 \\
-0.125 \\
0.375 \\
0.875 \\
1.375 \\
0.58 \\
-0.58 \\
-1.75 \\
-2.92 \\
-16.95 \\
-8.76 \\
-0.58 \\
7.61 \\
15.80 \\
\end{array} \]

p_z-like v-band wave fct.
Correction term large, dipole matrix element small

Global maximum magnitude of dipole matrix element: 0.36 Å
Distance between maximum of s-like and p\textsubscript{z}-like wave function: 0.71 Å
Scaled momentum matrix element $p_{cv}/m\omega_{cv}$: 5.81 Å
Lattice constant: 5.65 Å
Intersubband (THz) transitions in superlattices

Thin barrier (40Å):
- Depending on $z_0$, $r$ or $C$ can be as large as $p$
- Dipole matrix element can change sign
- Correction factor can change sign
- There exist zero-crossing of $C$

Thick barrier (120Å):
- Large region of very small $C$
- If cell boundary in barrier, $C$ negligible (this is often used as intuitive choice for unit cell)

$k=0$ wave functions, non-zero barrier thickness, left cell boundary at $z_0$
Nano-structures (e.g. quantum wells, dots): vanishing boundary conditions

Convenient "zone center approximation":

\[ u_{c,k}(r) = u_{c,0}(r), \quad u_{v,k}(r) = u_{v,0}(r) \]

\[ \int d^3r \, \Psi^*_{c,\ell}(r) \cdot r \cdot \Psi_{v,\ell}(r) \approx 0 \]

Compare Burt, 1993
Alternative proof in Gu et al, 2013

Generalized zone center approximation for nano-structures

\[ \Psi^{(nano)}_{v,\ell}(z) = \int_{BZ} d^3k \frac{\xi_{v,\ell}(k)}{(2\pi)^3} \, e^{i\vec{k}\cdot\vec{r}} \, u_{v,k}(r) \]

\[ \left< c, \ell' | r | v, \ell \right>_{\text{zone center}} \left< c, \ell' | i\vec{\nabla}_k | v, \ell \right>_{\text{cell}} \bigg|_{k=0} \int_{\text{all space}} d^3r \, \xi^*_{c,\ell'}(r) \cdot \xi_{v,\ell}(r) \]

Cell-envelope factorization involves \( k \)-gradient operator, not dipole operator

p-r relation for nano-structures in generalized zone-center approximation:

\[ \left< \Psi^{(nano,zca)}_{c,\ell'} | \hat{P} | \Psi^{(nano,zca)}_{v,\ell} \right> = \frac{im}{\hbar} \left( \epsilon_{c,0} - \epsilon_{v,0} \right) \left< c, \ell' | r | v, \ell \right>_{\text{zone center}} \]

no correction term
We noticed that, in the literature, $p-r$ relation for periodic boundary conditions is usually not correct (or defined ambiguously).

We generalized Yafet's correction term to the case of periodic boundary conditions.

For bulk GaAs, correction term found to be large for any location of unit cell.

For intersubband (THz) transitions in superlattices, correction term found to be small if barrier is wide and cell boundary is inside barrier.

We provided alternative proof to Blount's findings. This leads to vanishing correction term in infinite crystals but $p-r$ relation is in distribution sense and dipole operator is replaced by $k$-gradient operator.

For nano-structures, we developed alternative proof to Burt's finding that dipole matrix element essentially vanishes within zone-center approximation.

We showed that, for nano-structures, $p-r$ relation admits cell-envelope factorization, but dipole matrix element replaced by $k$-gradient matrix element.

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