Relation between Interband Dipole and Momentum Matrix Elements

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Reference: B. Gu, N.H. Kwong, R. Binder, Phys. Rev. B. **87**, 125301 (2013).

Infinite Volume Non-Vanishing Boundary Conditions	Finite Volume Periodic Boundary Conditions
$\langle c\,\mathbf{k}'\,\vert\,\mathbf{r}\,\vert\,v\,\mathbf{k}\rangle_{\infty} = \delta(\mathbf{k}-\mathbf{k}')\langle u_{c\,\mathbf{k}'}\,\vert\, i\vec{\nabla}_{\mathbf{k}}\,\vert\, u_{v\,\mathbf{k}}\rangle_{\text{cell}}$ Blount 1962	$\langle c\,\mathbf{k}\,\vert\,\mathbf{r}\,\vert\,v\,\mathbf{k}\rangle_{\text{vol}} \neq \langle u_{c\,\mathbf{k}}\,\vert\, i\vec{\nabla}_{\mathbf{k}}\,\vert\, u_{v\,\mathbf{k}}\rangle_{\text{cell}}$ Incorrectly proven in Haug 1972
$\langle u_{c\,\mathbf{k}}\,\vert\,\hat{\mathbf{p}}\,\vert\, u_{v\,\mathbf{k}}\rangle_{\text{cell}} = im\omega_{c\,\mathbf{k},v\,\mathbf{k}}\langle u_{c\,\mathbf{k}}\,\vert\, i\vec{\nabla}_{\mathbf{k}}\,\vert\, u_{v\,\mathbf{k}}\rangle_{\text{cell}}$ Adams 1952, Haug 1972	$\langle c\,\mathbf{k}\,\vert\,\hat{\mathbf{p}}\,\vert\,v\,\mathbf{k}\rangle_{\text{vol}} = im\omega_{c\,\mathbf{k},v\,\mathbf{k}}\langle u_{c\,\mathbf{k}}\,\vert\, i\vec{\nabla}_{\mathbf{k}}\,\vert\, u_{v\,\mathbf{k}}\rangle_{\text{cell}}$ Haug 1972
$\langle u_{c\,\mathbf{k}}\,\vert\,\hat{\mathbf{p}}\,\vert\, u_{v\,\mathbf{k}}\rangle_{\text{cell}} = im\omega_{c\,\mathbf{k},v\,\mathbf{k}}\langle u_{c\,\mathbf{k}}\,\vert\,\mathbf{r}\,\vert\, u_{v\,\mathbf{k}}\rangle_{\text{cell}} + \mathbf{B}_{c\,v}(\mathbf{k})$ Yafet 1957, Peeters etal. 1993, Foreman 2000	
$\langle c\,\mathbf{k}'\,\vert\,\hat{\mathbf{p}}\,\vert\,v\,\mathbf{k}\rangle_{\infty} = im\omega_{c\,\mathbf{k}',v\,\mathbf{k}}\langle c\,\mathbf{k}'\,\vert\,\mathbf{r}\,\vert\,v\,\mathbf{k}\rangle_{\infty}$ Follows from above, see Gu et al. 2013	$\langle c\,\mathbf{k}'\,\vert\,\hat{\mathbf{p}}\,\vert\,v\,\mathbf{k}\rangle_{\text{vol}} = im\omega_{c\,\mathbf{k}',v\,\mathbf{k}}\langle c\,\mathbf{k}'\,\vert\,\mathbf{r}\,\vert\,v\,\mathbf{k}\rangle_{\text{vol}} + \mathbf{C}_{c\,\mathbf{k}',v\,\mathbf{k}}$ This work

$$\hat{\mathbf{p}} = \frac{im}{\hbar}\left[\mathrm{H}_0, \mathbf{r}\right]_-$$

(classically: p=mv)

$$H_0 = -\frac{\hbar^2}{2m}\nabla^2 + V_L(\boldsymbol{r})$$

In crystals with periodic boundary conditions:

$$\langle \mathrm{c}\,\mathbf{k}'|\hat{\mathbf{p}}|\mathrm{v}\,\mathbf{k}\rangle_{\mathrm{vol}} = \frac{im}{\hbar}\langle \mathrm{c}\,\mathbf{k}'|H_0\mathbf{r} - \mathbf{r}H_0|\mathrm{v}\,\mathbf{k}\rangle_{\mathrm{vol}}$$

$$\langle \mathrm{c}\,\mathbf{k}'|\mathbf{r}H_0|\mathrm{v}\,\mathbf{k}\rangle_{\mathrm{vol}} = \varepsilon_{\mathrm{v}\,\mathbf{k}}\langle \mathrm{c}\,\mathbf{k}'|\mathbf{r}|\mathrm{v}\,\mathbf{k}\rangle_{\mathrm{vol}}$$

$$\langle \mathrm{c}\,\mathbf{k}'|H_0\mathbf{r}|\mathrm{v}\,\mathbf{k}\rangle_{\mathrm{vol}} \neq \varepsilon_{c\,\mathbf{k}}\langle \mathrm{c}\,\mathbf{k}'|\mathbf{r}|\mathrm{v}\,\mathbf{k}\rangle_{\mathrm{vol}}$$

not equal

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

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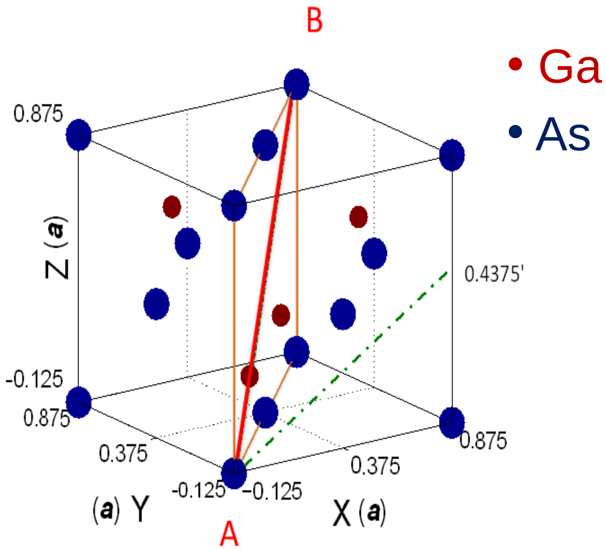
Comparison: infinite system, non-vanishing boundary conditions

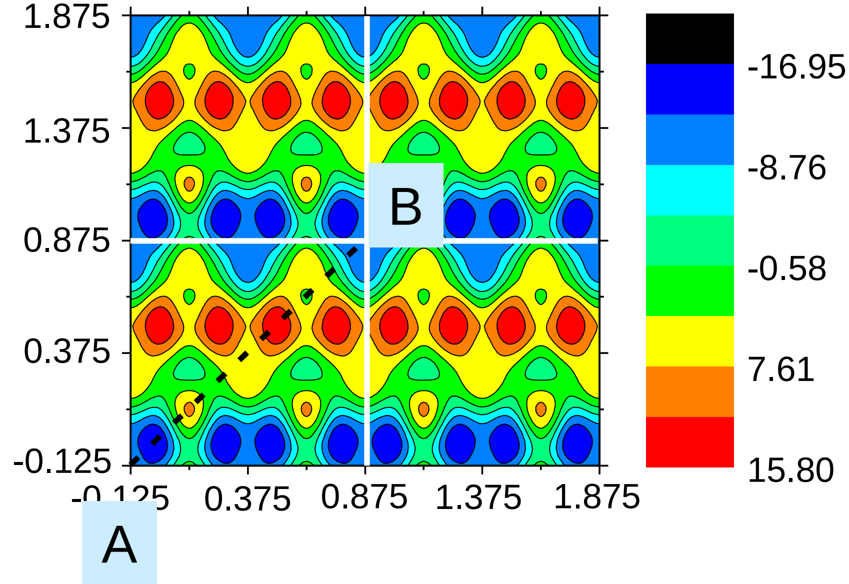
$$\langle \mathrm{c}\,\mathbf{k}'|\hat{\mathbf{p}}|\mathrm{v}\,\mathbf{k}\rangle_{\infty} = im\omega_{\mathrm{c}\,\mathbf{k}',\mathrm{v}\,\mathbf{k}}\langle \mathrm{c}\,\mathbf{k}'|\mathbf{r}|\mathrm{v}\,\mathbf{k}\rangle_{\infty}$$

- 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 $\mathbf{k}=\mathbf{k}'$ not defined
- Proof using limiting procedure with spatially limited wave-packets in Gu et al. 2013

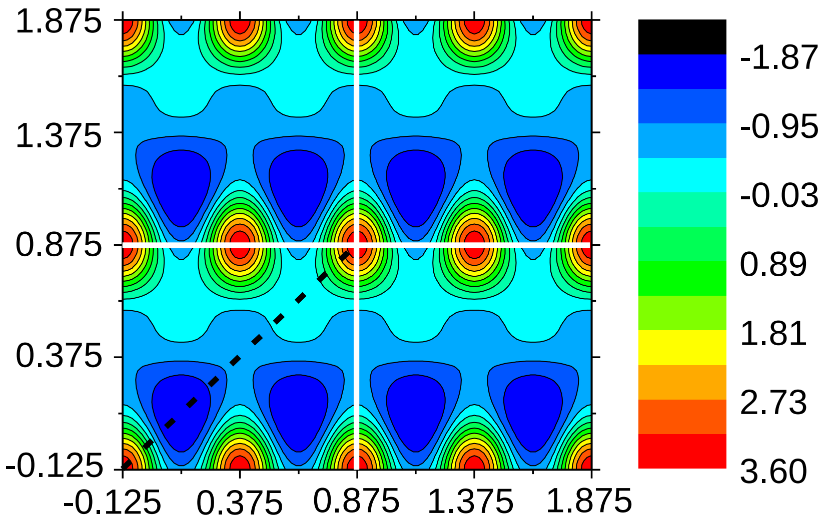
Bulk GaAs: simple Cohen-Bergstresser pseudopotential approach



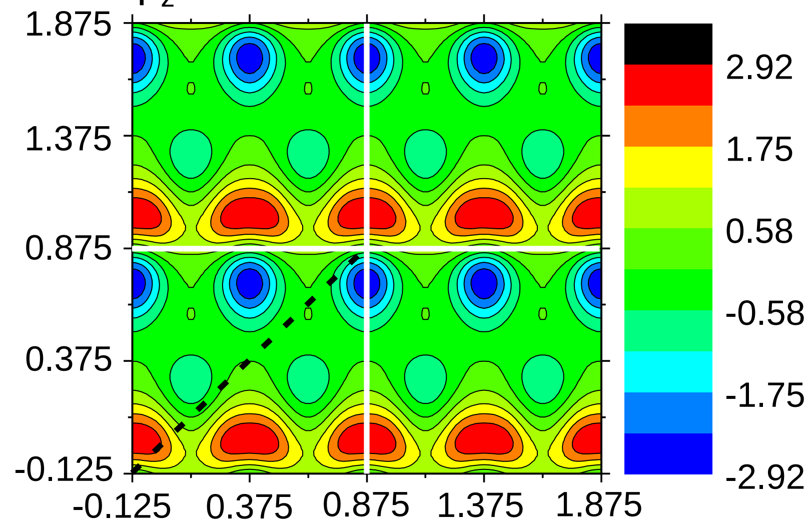
Pseudopotential:

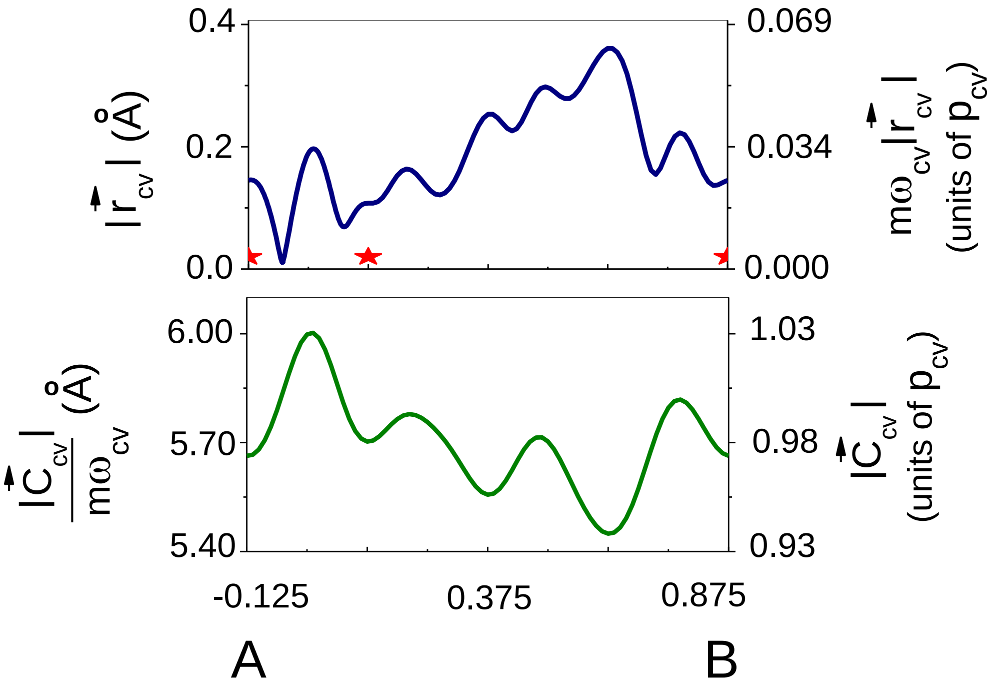


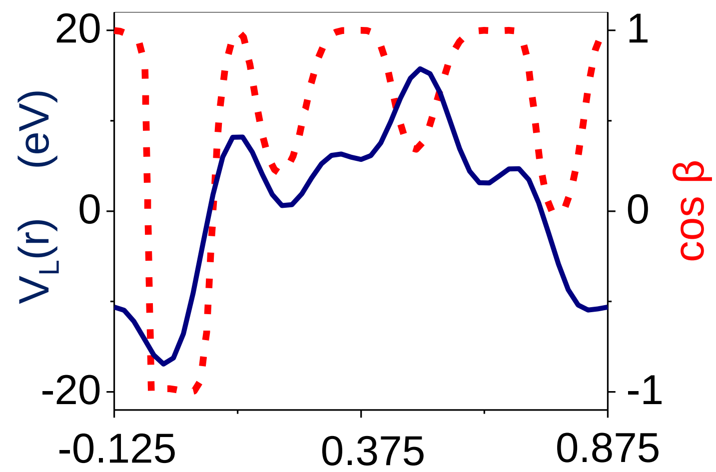
s-like c-band wave fct.

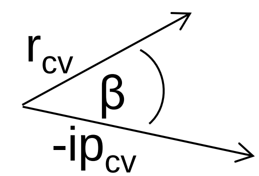


p_z-like v-band wave fct.





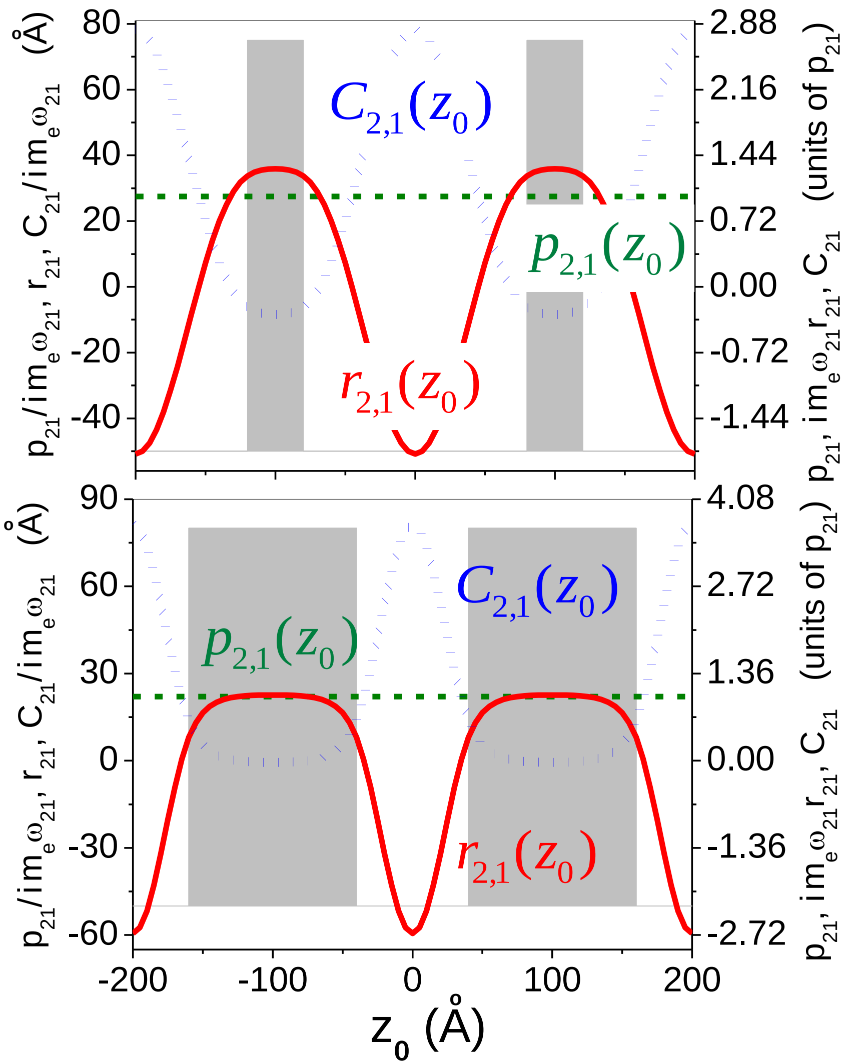




Correction term large, dipole matrix element small

Global maximum magnitude of dipole matrix element:	0.36 Å
Distance between maximum of s-like and p_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

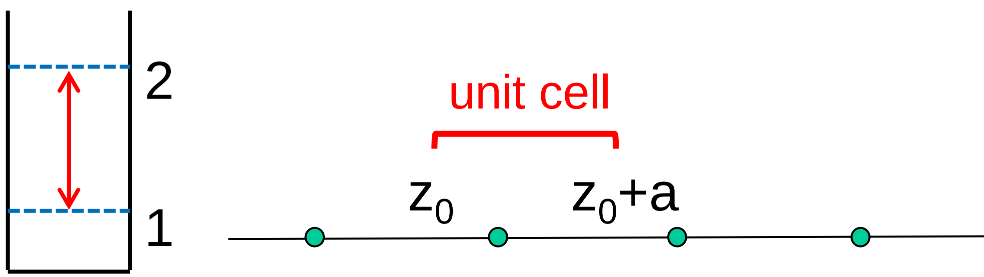


<u>Thin barrier (40Å):</u>

- ❑ Depending on z_0, r or C an be as large as p
- ❑ Dipole matrix element can change sign
- ❑ Correction factor can change sign
- ❑ There exist zero-crossing of C

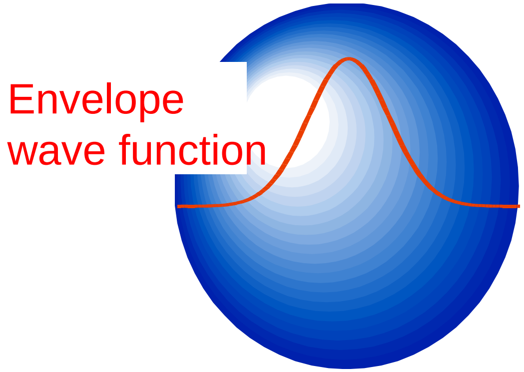
<u>Thick barrier (120Å):</u>

- ❑ 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



quantum dot

Convenient "zone center approximation":

$$u_{\mathrm{c},\boldsymbol{k}}(\boldsymbol{r}) = u_{\mathrm{c},\boldsymbol{0}}(\boldsymbol{r}), \quad u_{\mathrm{v},\boldsymbol{k}}(\boldsymbol{r}) = u_{\mathrm{v},\boldsymbol{0}}(\boldsymbol{r})$$

$$\int_{\text{qu. dot}} d^3r\, \Psi^*_{\mathrm{c}\ell'}(\boldsymbol{r})\, \boldsymbol{r}\, \Psi_{\mathrm{v}\ell}(\boldsymbol{r}) \simeq \boldsymbol{0}$$

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

Generalized zone center approximation for nano-structures

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

$$\langle \mathrm{c}\ell' | \boldsymbol{r} | \mathrm{v}\ell \rangle^{\text{zone center}}_{\text{nano}} = \langle \mathrm{u}_{\mathrm{c}\,\boldsymbol{k}} | i\vec{\nabla}_{\boldsymbol{k}} | \mathrm{u}_{\mathrm{v}\,\boldsymbol{k}} \rangle_{\text{cell}} \Big|_{\boldsymbol{k}=0} \int_{\text{all space}} d^3r\, \xi^*_{\mathrm{c}\ell'}(\boldsymbol{r})\, \xi_{\mathrm{v}\ell}(\boldsymbol{r})$$

Cell-envelope factorization involves k-gradient operator, not dipole operator

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

$$\left\langle \Psi^{(\mathrm{nano,zca})}_{\mathrm{c}\ell'} \middle| \hat{\mathbf{p}} \middle| \Psi^{(\mathrm{nano,zca})}_{\mathrm{v}\ell'} \right\rangle = \frac{im}{\hbar}(\varepsilon_{\mathrm{c},\mathbf{0}} - \varepsilon_{\mathrm{v},\mathbf{0}}) \langle \mathrm{c}\,\ell' | \mathbf{r} | \mathrm{v}\,\ell \rangle^{\text{zone center}}_{\text{nano}}$$

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 bound. 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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