Høyere ordens elektron-fonon-kobling

$$H_{el\text{-}ion} = \sum_i U(\vec{r}_i) = \sum_{ij} V_{el\text{-}ion}(\vec{r}_i - \vec{R}_j)$$

$$\vec{R}_j = R_j^0 - \sum_\lambda \vec{X}_{j\lambda}$$ Avvik fra likevekt.

λ: Indekser svingemode.

$$V_{el\text{-}ion}(\vec{r}_i - \vec{R}_j)$$

$$= V_{el\text{-}ion}(\vec{r}_i - \vec{R}_j^0)$$

Kontraksjon av to vektorer $\underbrace{X_{i\lambda} \ F_i}$

$$+ \sum_\lambda \underbrace{\vec{X}_{j\lambda}}_{\substack{\text{Vektor} \\ X_{i\lambda}}} \odot \underbrace{\vec{\nabla} V_{el\text{-}ion}}_{\substack{\text{Vektor} \\ F_i}} \Big|_{\vec{r}_i - R_j^0}$$

$$+ \frac{1}{2!} \sum_{\lambda_1} \sum_{\lambda_2} \underbrace{\vec{X}_{j\lambda_1} \vec{X}_{j\lambda_2}}_{\substack{\text{2. ordens} \\ \text{tensor} \\ A_{ij}}} \ \vdots \ \overbrace{\underbrace{\vec{\nabla}\vec{\nabla} V_{el\text{-}ion}}_{\substack{\text{2. ordens} \\ \text{tensor}}}}^{B_{ln}} \Big|_{\vec{r}_i - \vec{R}_j^0} + \cdots$$

Kontraksjon av to 2. ordens tensor

$$\underbrace{A_{ij} \, B_{ji}}$$

$$V_{el-ion}(\vec{r}) = \sum_{\vec{q}} \tilde{V}_{e-i}(\vec{q})\, e^{i\vec{q}\cdot\vec{r}}$$

$$\vec{\nabla}\vec{\nabla}\, V_{el-ion}(\vec{r})$$

$$= -\sum_{\vec{q}} \underbrace{\vec{q}\,\vec{q}}_{\text{tensor}}\; \tilde{V}_{e-i}(\vec{q})\, e^{i\vec{q}\cdot\vec{r}}$$

Dette leddet gir dermed følgende bidrag til koblingen mellom gittervibrasjoner og elektroner:

$$H^{(3)}_{el-ion} = -\frac{1}{2!}\sum_{i}\sum_{j,\lambda_1}\sum_{\lambda_2} \vec{X}_{j,\lambda_1}\,\vec{X}_{j,\lambda_2} : \sum_{\vec{q}} \vec{q}\,\vec{q}\;\tilde{V}_{e-i}(\vec{q})\, e^{i\vec{q}\cdot(\vec{r}_i - \vec{R}_j^{\,0})}$$

$$= -\frac{1}{2!}\sum_{i}\sum_{\substack{\vec{q}\\ \lambda_1\lambda_2}} \left\{ \sum_{j} \vec{X}_{j\lambda_1}\,\vec{X}_{j\lambda_2}\; e^{-iq\vec{R}_j^{\,0}} \right\} \vec{q}\,\vec{q}\;\tilde{V}_{e-i}(\vec{q})\, e^{i\vec{q}\cdot\vec{r}_i}$$

$$\vec{X}_{j\lambda} = \sum_{\vec{q}} \vec{X}_{q\lambda} e^{i\vec{q}\cdot\vec{R}_j^0} \Rightarrow$$

$$\sum_j \vec{X}_{j\lambda_1} \vec{X}_{j\lambda_2} e^{-iq\vec{R}_j^0}$$

$$= \sum_j \sum_{\vec{q}_1} \sum_{\vec{q}_2} \vec{X}_{q_1\lambda_1} \vec{X}_{q_2\lambda_2} e^{i(\vec{q}_1+\vec{q}_2-\vec{q})\cdot\vec{R}_j^0}$$

$$= \sum_{\vec{q}_1} \vec{X}_{q_1\lambda_1} \vec{X}_{\cancel{q}\vec{q}_1\lambda_2} = \sum_{\vec{q}'} \vec{X}_{\vec{q}'\lambda_1} \vec{X}_{q-\vec{q}',\lambda_2}$$

$$= \underbrace{\vec{\vec{H}}_{\vec{q}\lambda_1\lambda_2}}_{\text{2. ordens tensor.}}$$

$$H^{(2)}_{el-ion}(\vec{r}_i) = \sum_{\vec{q}} \tilde{T}(\vec{q}) e^{i\vec{q}\cdot\vec{r}_i}$$

$$H^{2}_{el-ion} = \sum_i H^{(2)}_{el-ion}(\vec{r}_i)$$

Da er 2. kvantisert form gitt ved

$$H^{2}_{el-ion} = \sum_{k,\vec{q},\sigma} \tilde{T}(\vec{q}) C^{+}_{k+\vec{q},\sigma} C_{k\sigma}$$

der $\tilde{T}(\vec{q})$ er Fourier-transformen til $H^{3}_{el-ion}(\vec{r})$.

$\tilde{T}(\vec{q})$

$$= \frac{-1}{2!} \sum_{\lambda_1 \lambda_2} \sum_{\vec{q}\,'} \left(\vec{X}_{q'\lambda_1} \; \vec{X}_{q-q',\lambda_2} : \vec{q}\,\vec{q} \right)$$

$$\tilde{V}_{e-i}(\vec{q})$$

Uttrykt ved kvantiserte gitter vibrasjoner, blir dette:

$$\vec{X}_{q'\lambda_1} \; \vec{X}_{q-q',\lambda_2}$$

$$= \frac{\hbar}{2M} \frac{1}{\sqrt{\omega_{q'\lambda_1} \, \omega_{q-q',\lambda_2}}} \left(a^+_{-q'\lambda_1} + a_{q'\lambda_1} \right) \cdot \left(a^+_{-(q-q')\lambda_2} + a_{q-q',\lambda_2} \right)$$

$$\left(\vec{\xi}_{q'\lambda_1} \; \vec{\xi}_{q-q'\lambda_2} : \vec{q}\,\vec{q} \right)$$

Dette er et langt svakere bidrag til elektron-fonon koblingen, enn det 1. ordens bidraget vi får. En viktig faktor til dette, er at $H^{(3)}_{el-ion}$ har en ekstra faktor $\sqrt{\hbar}$, er annen faktor som bidrar,

er at $H^{(1)}_{el-ion} \sim q$, mens

$$H^{(2)}_{el-ion} \sim q^2.$$

Dersom vi ser på akustiske langbølgede fononer (det er de som eksiteres først) har vi at bølgetallet $\vec{q}$ er lite, og dermed er også $q^2 \ll q$.

Vi multipliserer ut fonon-operatorene, slik at vi får:

$$\tilde{T}(\vec{q}) = \frac{-1}{2} \sum_{\lambda_1 \lambda_2} \sum_{\vec{q}\,'} \left(\vec{\xi}_{q'\lambda_1} \vec{\xi}_{\vec{q}-q',\lambda_2} : \vec{q}\,\vec{q} \right) \tilde{V}_{e-i}(\vec{q})$$

$$\frac{\hbar}{2M} \frac{1}{\sqrt{\omega_{q'\lambda_1}\, \omega_{q-q'\lambda_2}}}$$

$$\cdot \left(a^+_{-q'\lambda_1} a^+_{q'-q\,\lambda_2} + a^+_{-q'\lambda_1} a_{q-q'\lambda_2} + a_{q'\lambda_1} a^+_{q'-q,\lambda_2} + a_{q'\lambda_1} a_{q-q',\lambda_2} \right)$$

$$\cdot\; C^+_{k+q,\sigma}\, C_{k\sigma}$$

Dermed får vi, alt i alt:

$$H^{(3)}_{el-ion}$$

$$= \sum_{\substack{\vec{k},\vec{q},\vec{q}',\sigma \\ \lambda_1,\lambda_2}} S_{q,q',\lambda_1,\lambda_2} \cdot \left(a^{+}_{-q',\lambda_1} \cdot a^{+}_{q'-q,\lambda_2} + a^{+}_{-q',\lambda_1} a_{q-q',\lambda_2} + a_{q',\lambda_1} a^{+}_{q'-q,\lambda_2} + a_{q',\lambda_1} a_{q-q',\lambda_2} \right) \cdot C^{+}_{k+q,\sigma} C_{k,\sigma}$$

$$S = \frac{-1}{2!} \left[\vec{\xi}_{q',\lambda_1} \vec{\xi}_{q-q',\lambda_2} : \vec{q}\,\vec{q} \right] \frac{\hbar}{2M} \frac{1}{\sqrt{\omega_{q'\lambda_1} \; \omega_{q-q'\lambda_2}}} \tilde{V}_{e-i}(\vec{q})$$

Sammenligne dette med 1. orden kobling:

$$M_{q\lambda} = \Big[i \underbrace{\sqrt{\frac{\hbar}{2M\omega_{q\lambda}}}\;\vec{q}}_{\equiv \vec{A}_{q\lambda}} \cdot \vec{\xi}_{q\lambda} \Big] \tilde{V}_{e-i}(\vec{q})$$

$$M_{q\lambda} = \left[i \, \vec{A}_{q\lambda} \cdot \vec{\xi}_{q\lambda} \right] \tilde{V}_{e-i}(\vec{q})$$

$$S_{qq'\lambda\lambda'} = \left[\frac{i^2}{2!} \vec{A}_{q'\lambda} \vec{A}_{q-q'\lambda'} : \vec{\xi}_{q'\lambda} \vec{\xi}_{q-q'\lambda'} \right] \tilde{V}_{e-i}(\vec{q})$$

3. ordens leddet blir

$$W_{q q' q'' \lambda_1 \lambda_2 \lambda_3}$$

$$= \left[\frac{i^3}{3!} \vec{A}_{q'\lambda_1} \vec{A}_{q''\lambda_2} \vec{A}_{q-q'-q'',\lambda_3} \vdots \vec{\xi}_{q'\lambda_1} \vec{\xi}_{q''\lambda_2} \vec{\xi}_{q-q'-q'',\lambda_3} \right]$$

$$\tilde{V}_{e-i}(\vec{q})$$

etc.

Hver orden gir en faktor $\underline{\vec{q}\sqrt{\frac{\hbar}{2M}}}$

Til n'te orden har vi en kontraksjon av 2 n-te ordens tensorer

$$\underbrace{\vec{A} \cdots \vec{A}}_{n\text{-te ordens tensor}} \quad \text{og} \quad \underbrace{\vec{\xi} \cdots \vec{\xi}}_{n\text{-te ordens tensor.}}$$

Impulsbevarelse!

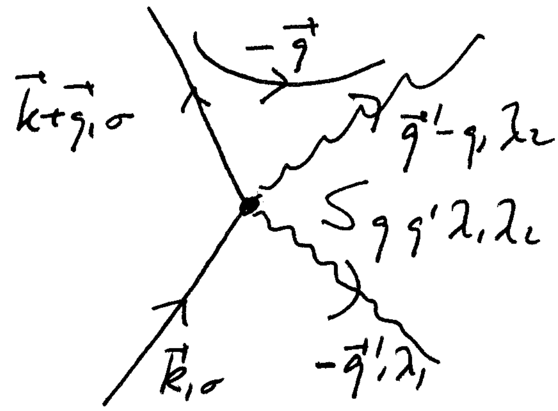
$$\vec{q}' + \vec{q}'' + \cdot \; q^{(n+1)} = \vec{q}$$

Fordel $\vec{q}', \vec{q}'' \cdots q^{(n-1)}$ på de $n-1$ første faktorene $\vec{A}_{q'} \cdots \vec{A}_{q^{(n-1)}}$. I den siste $\vec{A}$-faktoren: $\vec{A}_{q-q'-\cdots q^{(n)}}$

Tilsvarende på $\vec{\xi}$-faktorene.

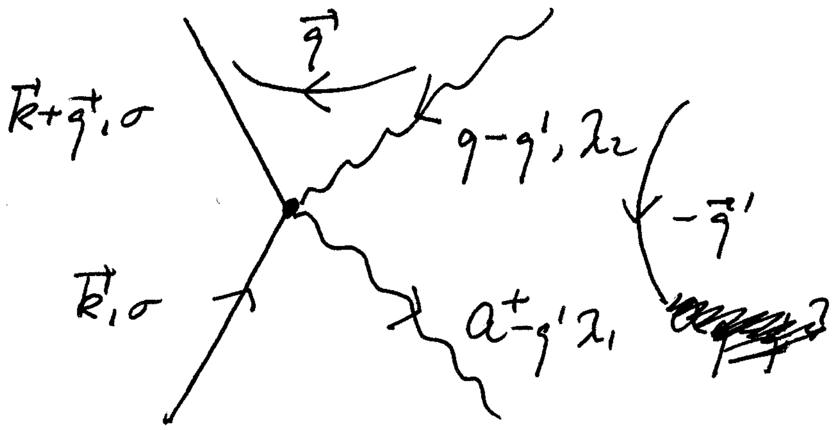
Vi får dermed 4 typer spredningsprosesser:

i)

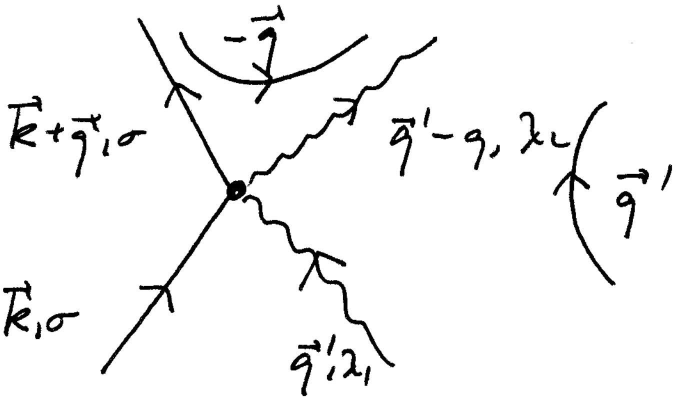


Impuls-konserv.

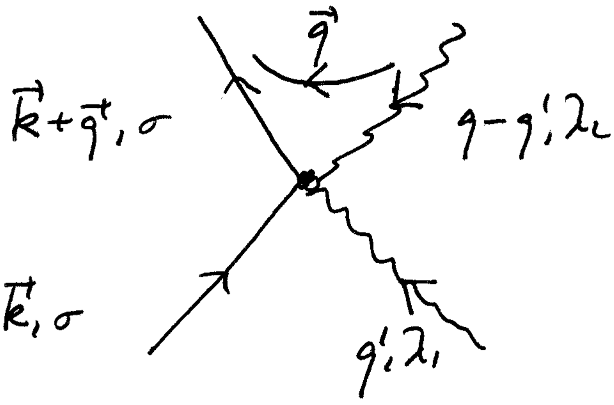
ii)



iii)



iv)



Vi gjennomfører også beregningen til 3. orden ⑨

3. ordens leddet: $V^{(3)}_{el-ion}(\vec{r}_i - \vec{R}_j^{\,0})$

$$= \frac{1}{3!}\sum_{\lambda_1\lambda_2\lambda_3} \underbrace{\vec{x}_{j\lambda_1}\,\vec{x}_{j\lambda_2}\,\vec{x}_{j\lambda_3}}_{\text{3. ordens tensor}} \vdots \underbrace{\vec{\nabla}\vec{\nabla}\vec{\nabla} V_{el-ion}(\vec{r}_i - \vec{R}_j^{\,0})}_{\text{3. ordens tensor}}$$

$$V_{el-ion}(\vec{r}) = \sum_{\vec{q}} \tilde{V}_{e-i}(\vec{q})\, e^{i\vec{q}\cdot\vec{r}}$$

$$\vec{\nabla}\vec{\nabla}\vec{\nabla} V_{el-ion}(\vec{r}) = i^3 \sum_{\vec{q}} \vec{q}\,\vec{q}\,\vec{q}\; e^{i\vec{q}\cdot\vec{r}}$$

$$x_{j\lambda} = \sum_{\vec{q}} \vec{X}_{q\lambda}\, e^{i\vec{q}\cdot\vec{R}_j^{\,0}}$$

Dermed får vi for $H^{(3)}_{el-ion}$:

$$H^{(3)}_{el-ion} = \sum_{i,j} \frac{1}{3!}\sum_{\lambda_1\lambda_2\lambda_3} \sum_{\vec{q}_1\vec{q}_2\vec{q}_3} \vec{X}_{q_1\lambda_1}\, \vec{X}_{q_2\lambda_2}\, \vec{X}_{q_3\lambda_3} \vdots\; i^3 \sum_{\vec{q}} \vec{q}\,\vec{q}\,\vec{q}\; e^{i\vec{q}\cdot(\vec{r}_i - \vec{R}_j^{\,0})} \cdot e^{i(\vec{q}_1+\vec{q}_2+\vec{q}_3)\cdot\vec{R}_j^{\,0}} \cdot \tilde{V}_{e-i}(\vec{q})$$

$$= \frac{i^3}{3!} \sum_i \sum_{\lambda_1 \lambda_2 \lambda_3} \sum_{\substack{\vec{q} \\ \vec{q}_1 \vec{q}_2 \vec{q}_3}} \left[\vec{X}_{q_1 \lambda_1} \vec{X}_{q_2 \lambda_2} \vec{X}_{q_3 \lambda_3} \vdots \vec{q}\, \vec{q}\, \vec{q} \right]$$

$$\cdot \underbrace{\sum_j e^{i(\vec{q}_1 + \vec{q}_2 + \vec{q}_3 - \vec{q}) \cdot R_j^0}}_{\delta_{q_1+q_2+q_3, q}}$$

$$\cdot\, e^{i \vec{q} \cdot \vec{r}_i} \quad \tilde{V}_{e-i}(\vec{q})$$

$$H^{(3)}_{el-ion}(\vec{r}_i)$$

$$= \frac{i^3}{3!} \sum_{\vec{q}} \tilde{G}(\vec{q})\, e^{i \vec{q} \cdot \vec{r}_i}$$

$$\tilde{G}(\vec{q}) = \frac{i^3}{3!} \sum_{\lambda_1 \lambda_2 \lambda_3} \sum_{q_1 q_2} \left[\vec{X}_{q_1 \lambda_1} \vec{X}_{q_2 \lambda_2} \vec{X}_{q - q_2 - q_3, \lambda_3} \vdots \vec{q}\, \vec{q}\, \vec{q} \right] \tilde{V}_{e-i}(\vec{q})$$

2. kvantisert form:

$$H^{(3)}_{el-im} = \sum_i H^{(3)}_{el-im}(\vec{r}_i)$$

$$= \sum_{k \vec{q} \sigma} \tilde{G}(\vec{q})\, C^{+}_{k+\vec{q}, \sigma} C_{k\sigma}$$

$$\vec{X}_{q\lambda} = \sqrt{\frac{\hbar}{2M\omega_{q\lambda}}}\, \vec{\xi}_{q\lambda}\, (a^+_{-q\lambda} + a_{q\lambda})$$

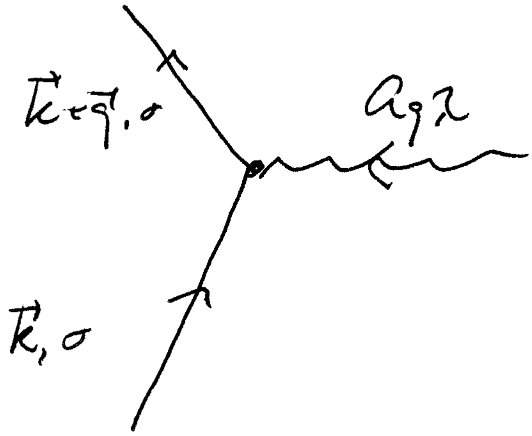
$$H^{(3)}_{el-ion} = \sum_{\substack{k, q, \sigma \\ q_1 q_2 \\ \lambda_1 \lambda_2 \lambda_3}} W_{q_1 q_2 q \lambda_1 \lambda_2 \lambda_3} \cdot (a^+_{-q_1 \lambda_1} + a_{q_1 \lambda_1}) \cdot (a^+_{-q_2 \lambda_2} + a_{q_2 \lambda_2}) \cdot (a^+_{-(q-q_2-q_3), \lambda_3} + a_{q-q_2-q_3, \lambda_3}) \cdot c^+_{k+q, \sigma}\, c_{k\sigma}$$

$$W_{q_1 q_2 q \lambda_1 \lambda_2 \lambda_3}$$

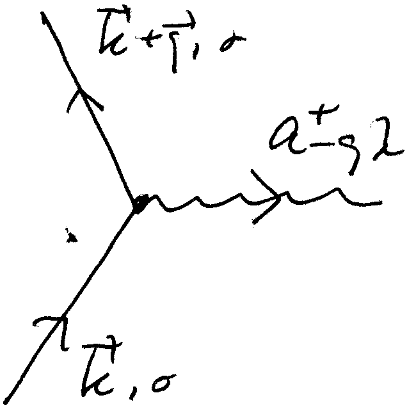
$$= \frac{i^3}{3!} \left(\frac{\hbar}{2M}\right)^{\frac{3}{2}} \frac{1}{\sqrt{\omega_{q_1\lambda_1}\, \omega_{q_2\lambda_2}\, \omega_{q-q_1-q_2, \lambda_3}}} \left[\vec{\xi}_{q_1\lambda_1}\, \vec{\xi}_{q_2\lambda_2}\, \vec{\xi}_{q-q_2-q_3, \lambda_3} \vdots \vec{q}\,\vec{q}\,\vec{q}\right] \tilde{V}_{e-i}(\vec{q})$$

$$= \frac{i^3}{3!} \left[\vec{A}_{q_1\lambda_1}\, \vec{A}_{q_2\lambda_2}\, \vec{A}_{q-q_1-q_2, \lambda_3} \vdots \vec{q}\,\vec{q}\,\vec{q}\right] \tilde{V}_{e-i}(\vec{q})$$

Et 1. ordens elektron-fonon koblings diagram ser slik ut:



Her treffer et fonon et elektron og sprer det



Her eksiterer et elektron en kvantisert gitter-vibrasjon.

Vi kan nå tenke oss at et elektron først eksiterer en gittervibrasjon, som så treffer et annet elektron. Dermed har vi effektivt fått en elektron-elektron v.v. via krystallgitterets eksitasjoner