Scattering from Harmonic Crystal [nln95]

Atoms of mass M are harmonically coupled via a bilinear form in displacement coordinates. Spatial Fourier transform produces normal modes: noninteracting collective excitations (phonons) representing oscillating patterns of specific wave vectors $\mathbf{k}$ and excitation energies determined by a characteristic dispersion relation $\epsilon(\mathbf{k})$.

$$\mathcal{H} = \sum_l \frac{p_l^2}{2M} + \frac{1}{2}\sum_{l,l'} \mathbf{u}_l \cdot \mathbf{D}_{ll'} \cdot \mathbf{u}_{l'} = \sum_{\mathbf{k}} \epsilon(\mathbf{k}) a_{\mathbf{k}}^\dagger a_{\mathbf{k}}.$$

Correlation function:[1]

$$\begin{aligned}\tilde{S}(\mathbf{q},t) &= \langle e^{-\imath\mathbf{q}\cdot\mathbf{u}_l} e^{\imath\mathbf{q}\cdot\mathbf{u}_{l'}(-t)}\rangle \\ &= \exp\left(-\frac{1}{2}\langle[\mathbf{q}\cdot\mathbf{u}_l]^2\rangle - \frac{1}{2}\langle[\mathbf{q}\cdot\mathbf{u}_{l'}(-t)]^2\rangle + \langle[\mathbf{q}\cdot\mathbf{u}_l][\mathbf{q}\cdot\mathbf{u}_{l'}(-t)]\rangle\right)\end{aligned}$$

Debye-Waller factor from $\frac{1}{2}\langle[\mathbf{q}\cdot\mathbf{u}_l]^2\rangle = \frac{1}{2}\langle[\mathbf{q}\cdot\mathbf{u}_{l'}(-t)]^2\rangle = W$.

Expansion into m-phonon processes:

$$\exp\Big(\langle[\mathbf{q}\cdot\mathbf{u}_l][\mathbf{q}\cdot\mathbf{u}_{l'}(-t)]\rangle\Big) = \sum_{m=0}^{\infty} \frac{1}{m!}\Big(\langle[\mathbf{q}\cdot\mathbf{u}_l][\mathbf{q}\cdot\mathbf{u}_{l'}(-t)]\rangle\Big)^m.$$

Dynamic structure factor:

$$S(\mathbf{q},\omega) = e^{-2W}\frac{1}{N}\sum_{ll'} e^{\imath\mathbf{q}\cdot(\mathbf{R}_l-\mathbf{R}_{l'})}\int_{-\infty}^{+\infty} dt e^{\imath\omega t}\exp\Big(\langle[\mathbf{q}\cdot\mathbf{u}_l][\mathbf{q}\cdot\mathbf{u}_{l'}(-t)]\rangle\Big).$$

$m = 0$: Bragg scattering

$$S(\mathbf{q},\omega)_0 \propto e^{-2W}\delta(\omega)\sum_{\mathbf{G}}\delta_{\mathbf{q},\mathbf{G}}; \quad \mathbf{G}: \text{ reciprocal lattice vector.}$$

$m = 1$: 1-phonon contributions[2]

$$S(\mathbf{q},\omega)_1 \propto e^{-2W}\frac{[\mathbf{q}\cdot\mathbf{e}(\mathbf{k})]^2}{2M\epsilon(\mathbf{k})}\Big(\underbrace{[1+n_{\mathrm{B}}(\mathbf{q})]\delta\big(\omega-\epsilon(\mathbf{q})\big)}_{\text{phonon emission}} + \underbrace{n_{\mathrm{B}}(\mathbf{q})\delta\big(\omega+\epsilon(\mathbf{q})\big)}_{\text{phonon absorption}}\Big).$$

Harmonicity leaves phonon peaks sharp. Thermal fluctuations only affect intensity via Debye-Waller factor.

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[1]Use $\langle e^A e^B\rangle = e^{\langle A^2+2AB+B^2\rangle/2}$ for operators A, B that are linear in $\mathbf{u}_l, \mathbf{p}_l$.

[2]Calculate $\langle[\mathbf{q}\cdot\mathbf{u}_\mathbf{0}][\mathbf{q}\cdot\mathbf{u}_\mathbf{R}(-t)]\rangle$ with $\mathbf{u}_\mathbf{R} \propto \sum_\mathbf{k}(2M\epsilon(\mathbf{k}))^{-1/2}(a_\mathbf{k}+a_\mathbf{k}^\dagger)e^{\imath\mathbf{k}\cdot\mathbf{R}}\mathbf{e}(\mathbf{k})$ and $a_\mathbf{k}(t) = a_\mathbf{k}e^{-\imath\epsilon(\mathbf{k})t}$.