CINS Summer School 2013

Basic Theory II
– Inelastic Scattering

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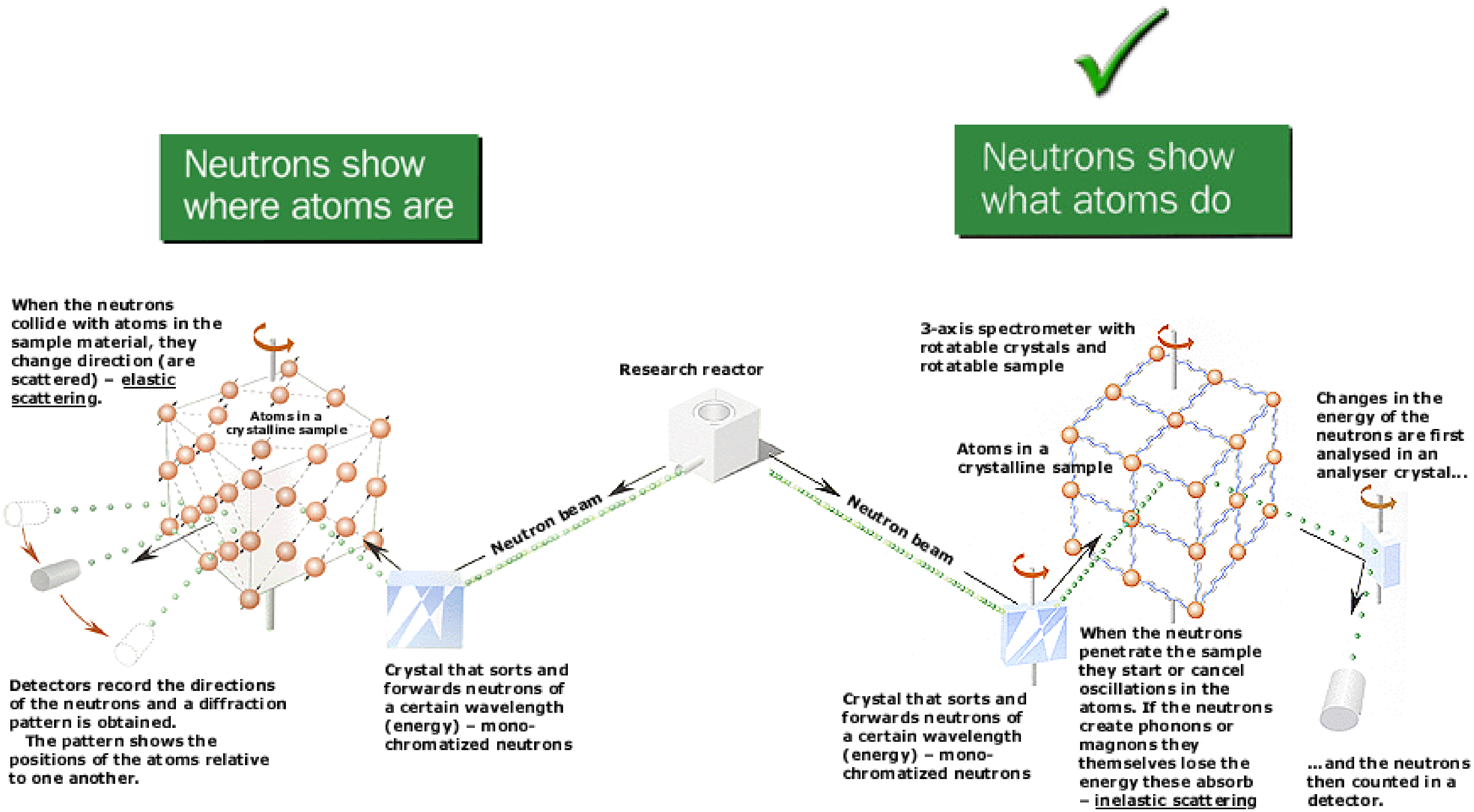
Outline

- Motivation – Inelastic Scattering
- Elementary excitations
  - Phonons
  - Magnons
- Practical aspects
- Scattering cross-section essentials
- One phonon cross-section
  - Example of Cu
- Magnons
- Damped harmonic oscillators

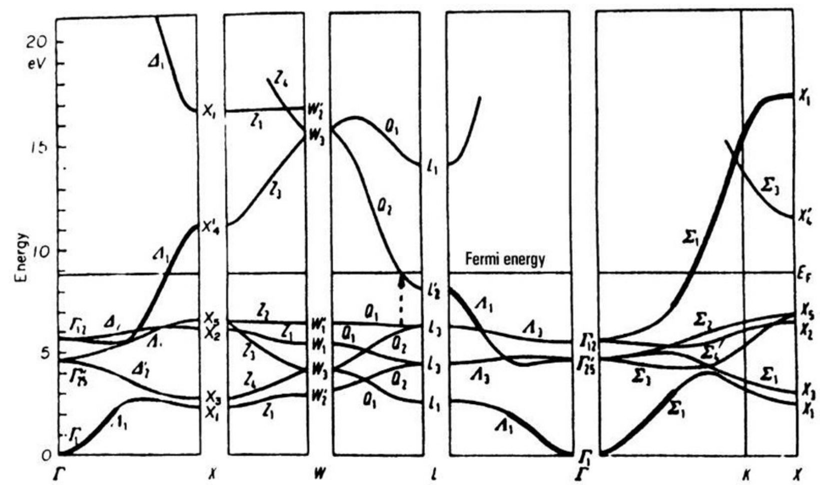
Inelastic Neutron Scattering

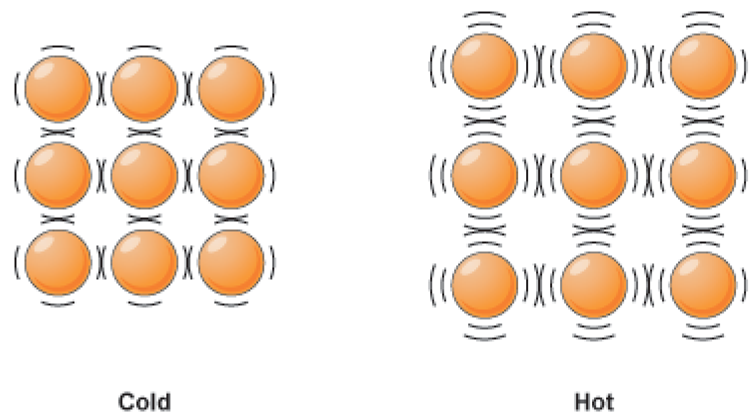


1994 Nobel prize poster (C. G. Shull and B. N. Brockhouse)

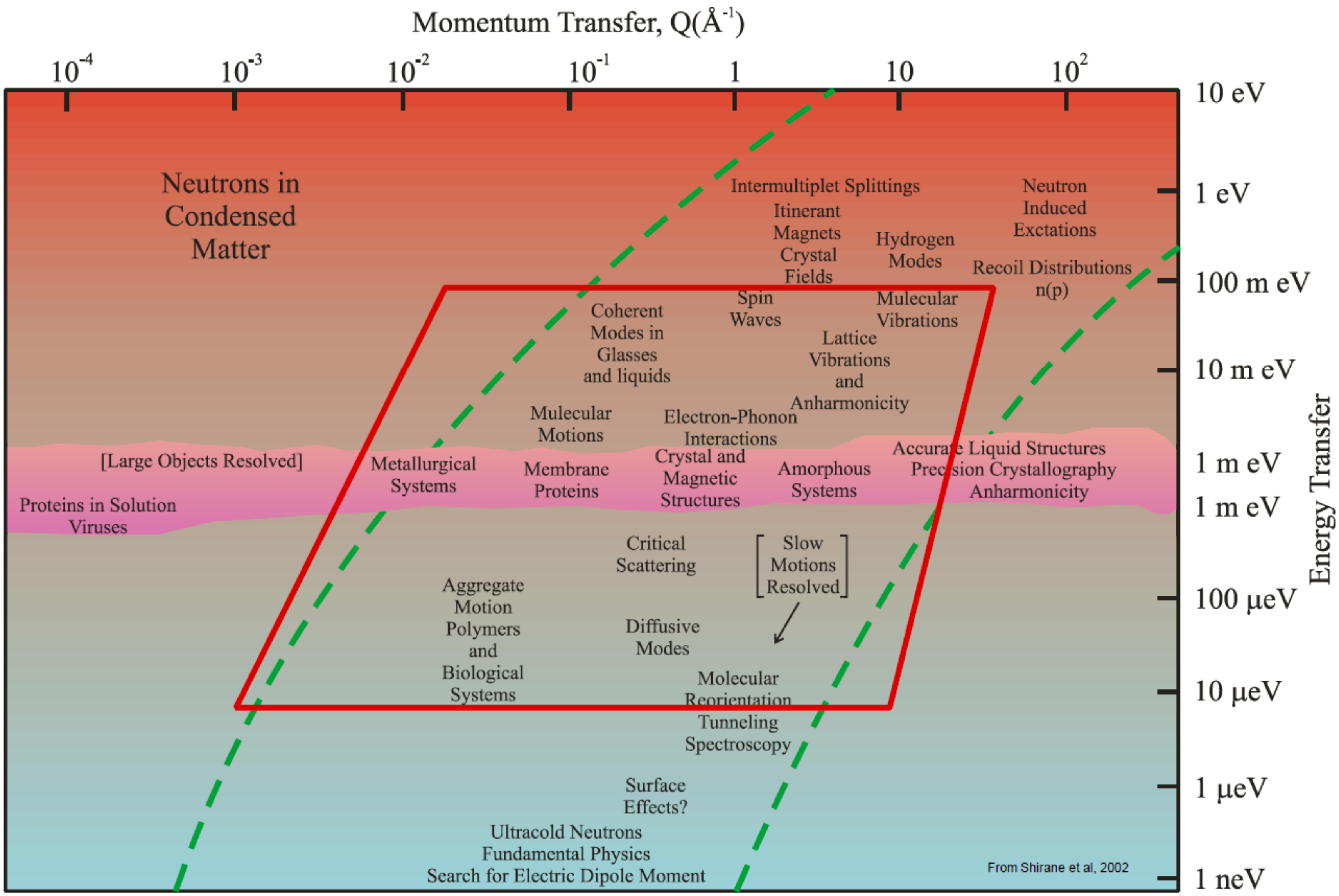
Excitations in solids

- Physical properties are determined by excitations, such as phonons and electronic excitations





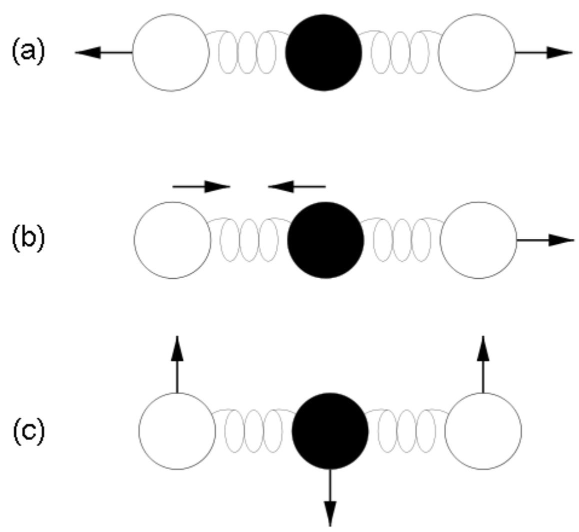
Inelastic Scattering Atlas



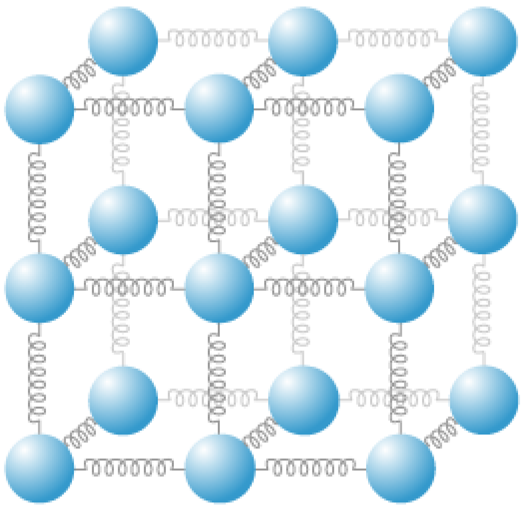
Phonon

- Propagating lattice vibration
- Carries most of entropy in a material
- Responsible for many thermodynamic phenomena (Specific heat, thermal conduction, etc.)

http://spark.ucar.edu/molecular-vibration-modes



molecules

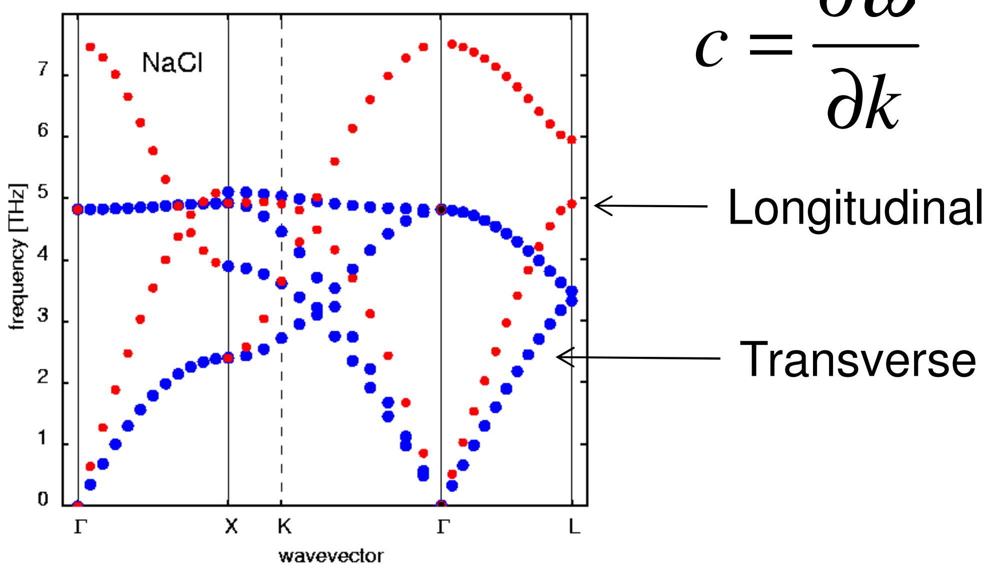


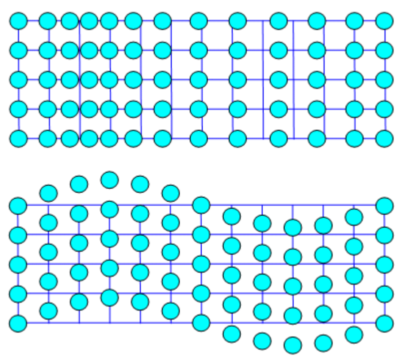
solid

Phonon propagation

Molecules/gas	Liquid/glass	Solid (crystal)
Vibrational and rotational modes Do not propagate – no momentum dependence		Propagating vibrational mode – both longitudinal and transverse mode e.g. earthquakes

$$c = \frac{\partial \omega}{\partial k}$$



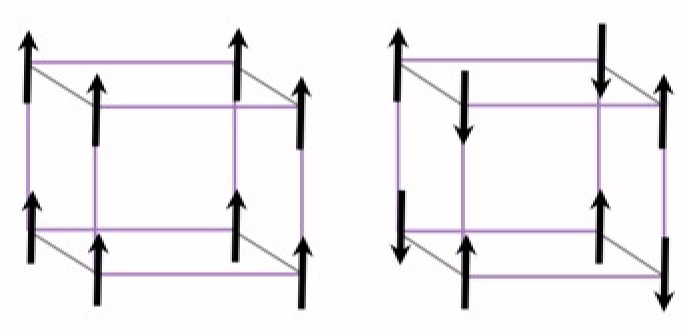


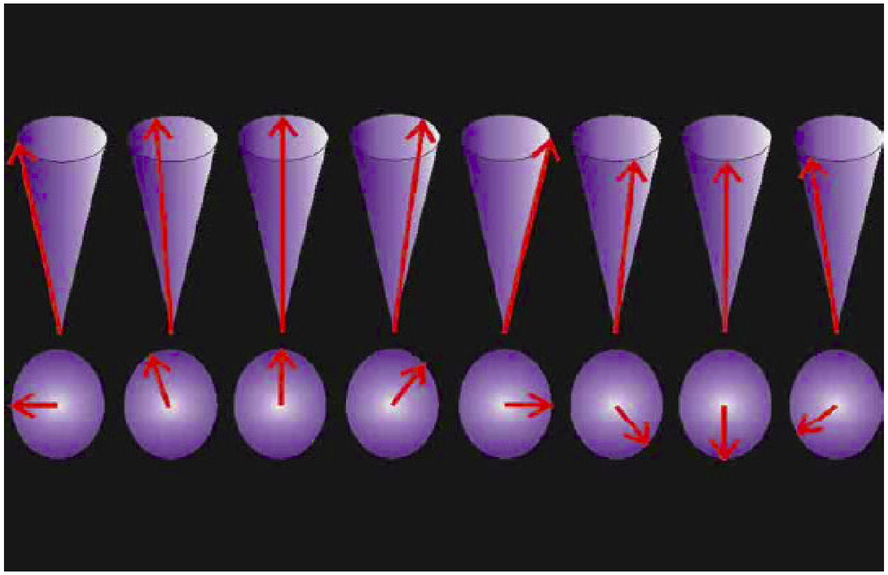
http://people.web.psi.ch/delley/nacl.html

http://www.tf.uni-kiel.de/matwis/amat/mw2_ge/kap_2/advanced/t2_1_1.html

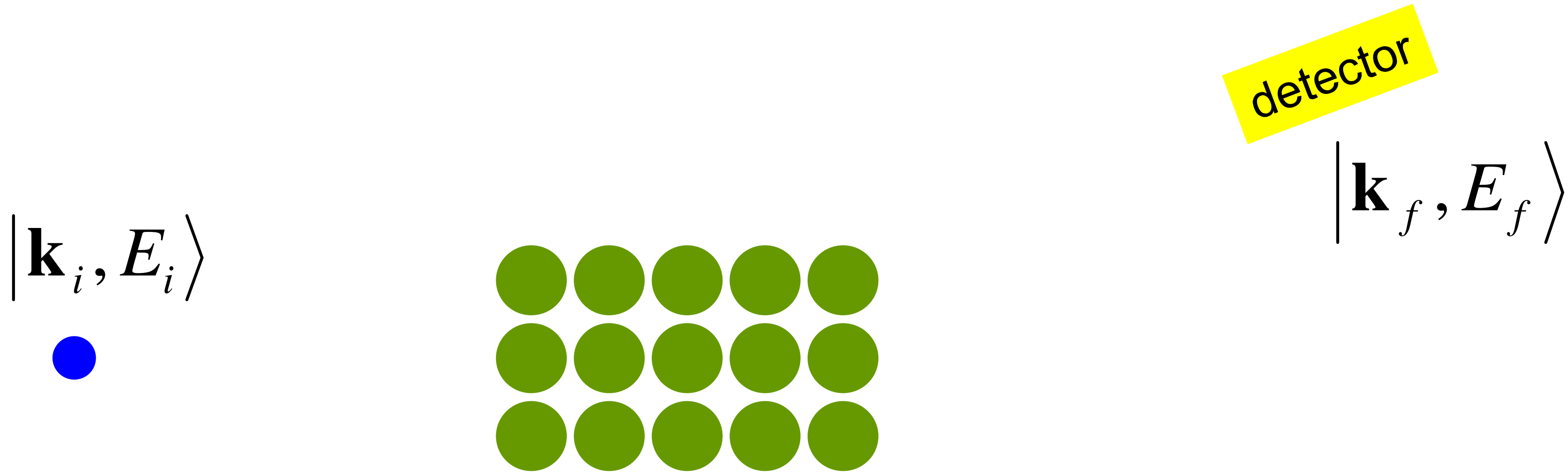
Magnon (spin wave)

- Magnetic order (spin solid)
  - Ferromagnetic or
  - Antiferromagnetic
- Magnetic moment fluctuations can propagate in the presence of order
  - Spin wave or magnon
  - Only transverse!
- In general, material's magnon dispersion is easier to calculate than its phonon counterpart. Why?





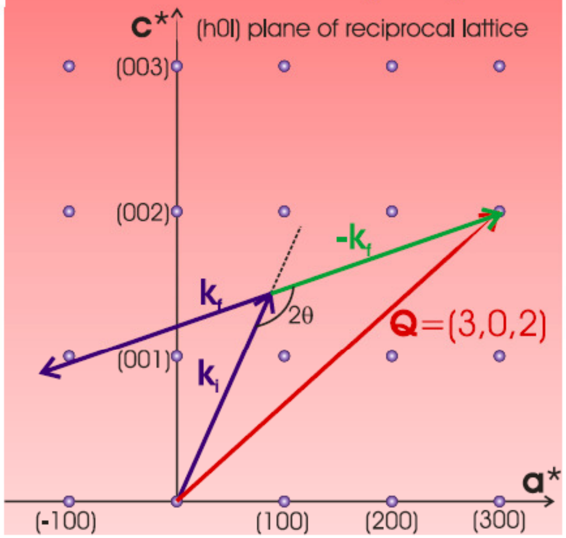
How to measure these in practice?



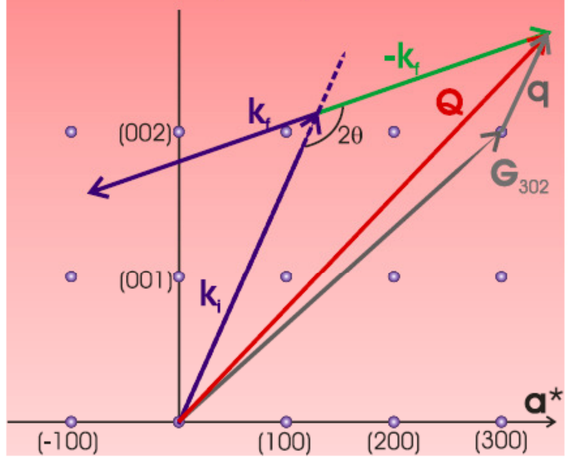
- Neutron scattering data ➔ Histogram of events
- Inelastic scattering events different from diffraction
- Poisson distribution
  - σ is given by $\sqrt{\#\text{ of events}}$

Scattering triangle

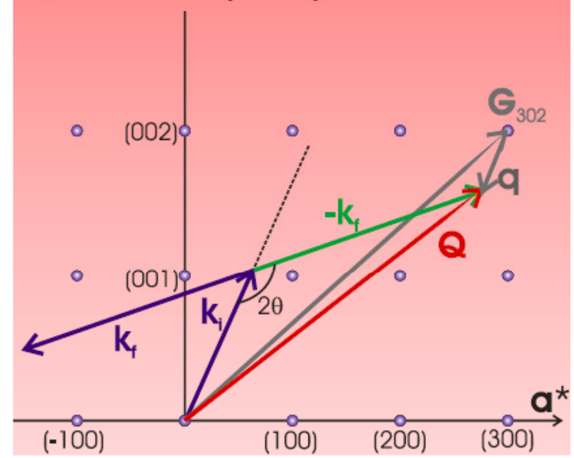
Elastic: $\mathbf{k}_i=\mathbf{k}_f$



Neutron Energy loss: $\mathbf{k}_f<\mathbf{k}_i$



Neutron Energy gain: $\mathbf{k}_f>\mathbf{k}_i$



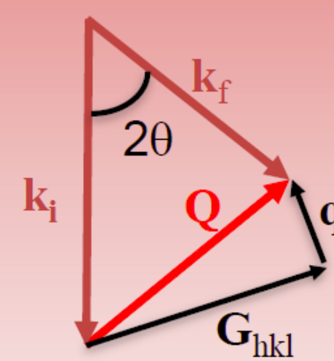
Kinematic range that can be covered in a scattering event:

$$\frac{\hbar^2}{2m}Q^2 = E_i + E_f - 2\sqrt{E_i E_f}\cos 2\theta$$

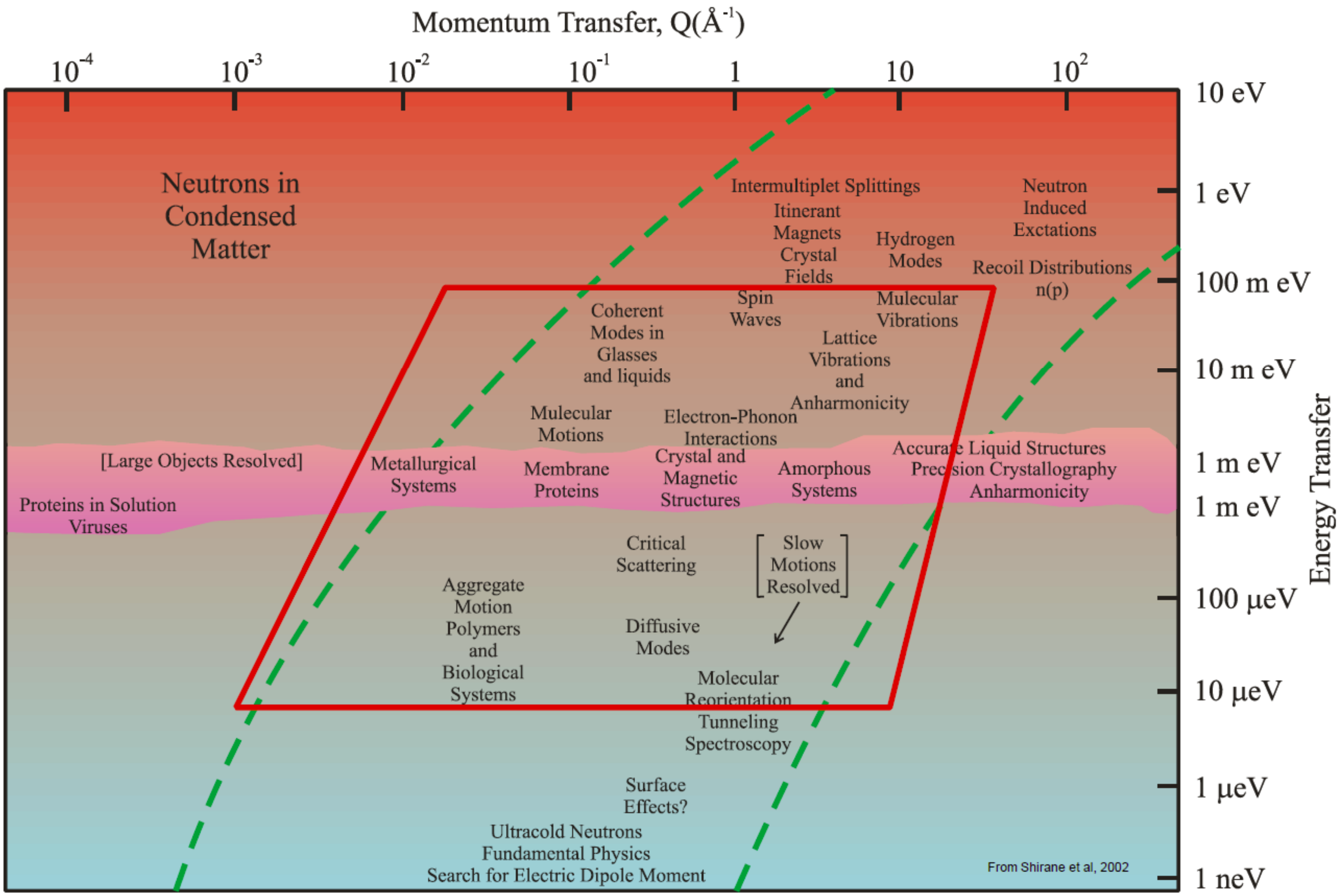
$$\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f = \mathbf{G}_{hkl} + \mathbf{q}$$

$$Q^2 = k_i^2 + k_f^2 - 2k_i k_f \cos 2\theta$$

$$E = E_i - E_f \rightarrow \frac{\hbar^2}{2m}Q^2 = 2E_f + E - 2\sqrt{E_f(E_f+E)}\cos 2\theta$$



Inelastic Scattering Atlas



Cross-section Essentials I

- Inelastic neutron scattering measures dynamic structure factor S(**Q**, ω):

$$S(\mathbf{Q},\omega) = \frac{1}{2\pi\hbar N}\sum_{j,j'}\int_{-\infty}^{\infty} dt \left\langle e^{+i\mathbf{Q}\cdot\mathbf{r}_j(t)} e^{-i\mathbf{Q}\cdot\mathbf{r}_{j'}(0)} \right\rangle e^{-i\omega t}$$

- Dynamic structure factor S(**Q**, ω) is the Fourier Transformation of space-time correlation function
- Dynamic structure factor S(**Q**, ω) is related to the imaginary part of the dynamic susceptibility χ(**Q**, ω) through the fluctuation-dissipation theorem

$$S(\mathbf{Q},\omega) = \frac{\chi''(\mathbf{Q},\omega)}{1 - e^{-\hbar\omega/k_B T}}$$

Cross-section Essentials II

- Coherent one phonon scattering:

$$S(\mathbf{Q},\omega) \propto \frac{(\mathbf{Q}\cdot\mathbf{e}_s)^2}{\omega_{\mathbf{q}s}}\left[\langle n_s+1\rangle\delta(\omega-\omega_{\mathbf{q}s})+\langle n_s\rangle\delta(\omega+\omega_{\mathbf{q}s})\right]$$

phonon polarization (eigenvector)

phonon dispersion relation

phonon annihilation

phonon creation

$$\langle n_s\rangle = \frac{1}{e^{\hbar\omega_{\mathbf{q}s}/k_B T}-1}$$

- Detailed balance: Probability of a transition depends on statistical weight factor of the initial state:

$$S(-\mathbf{Q},-\omega) = e^{-\hbar\omega/k_B T} S(\mathbf{Q},\omega)$$

Derivations

- Can be found in references:
  - Squires, *Introduction to the theory of thermal neutron scattering*
  - Shirane, Shapiro, and Tranquada, *Neutron scattering with a triple-axis spectrometer*
  - Marder, *Condensed matter physics*
  - Dove, Structure and Dynamics
  - Chaikin and Lubensky, *Principles of condensed matter physics*
- Do it yourself!
- I will talk about rough steps for one phonon cross-section

$$S(\mathbf{Q},\omega)=\frac{1}{2\pi\hbar N}\sum_{j,j'}\int_{-\infty}^{\infty}dt\left\langle e^{+i\mathbf{Q}\cdot\mathbf{r}_j(t)}e^{-i\mathbf{Q}\cdot\mathbf{r}_{j'}(0)}\right\rangle e^{-i\omega t}$$

$$\mathbf{r}_j(t)=\mathbf{R}_j+\mathbf{u}_j(t)$$

average position

instantaneous displacement (Harmonic)

$$\mathbf{u}_j=\frac{1}{\sqrt{N}}\sum_{\mathbf{k}\nu}\left[\hat{u}_{\mathbf{k}\nu}e^{i\mathbf{k}\cdot\mathbf{R}_j}+\hat{u}^{+}_{\mathbf{k}\nu}e^{-i\mathbf{k}\cdot\mathbf{R}_j}\right]\quad \hat{u}_{\mathbf{k}\nu}\equiv\sqrt{\frac{\hbar}{2M\omega_{\mathbf{k}\nu}}}\mathbf{e}_{\mathbf{k}\nu}\hat{a}_{\mathbf{k}\nu}$$

$$\sum_{j,j'}e^{i\mathbf{Q}\cdot(\mathbf{R}_j-\mathbf{R}_{j'})}\int_{-\infty}^{\infty}dt\left\langle e^{+i\mathbf{Q}\cdot(\mathbf{u}_j(t)-\mathbf{u}_{j'}(0))}\right\rangle e^{-i\omega t}$$

$$\left\langle e^{(A+B)}\right\rangle=e^{\left\langle\frac{(A+B)^2}{2}\right\rangle}$$

$$\langle\cdots\rangle=e^{-\left\langle\frac{[\mathbf{Q}\cdot\mathbf{u}_j(t)]^2}{2}\right\rangle+\left\langle[\mathbf{Q}\cdot\mathbf{u}_j(t)][\mathbf{Q}\cdot\mathbf{u}_{j'}(0)]\right\rangle-\left\langle\frac{[\mathbf{Q}\cdot\mathbf{u}_{j'}(0)]^2}{2}\right\rangle}$$

mean square ionic displacement (time indep.)

$$e^{-2W} = e^{-\left\langle (\mathbf{Q}\cdot\mathbf{u}_j)^2 \right\rangle} \quad : \text{Debye} - \text{Waller factor}$$

$$e^{\left\langle [\mathbf{Q}\cdot\mathbf{u}_j(t)][\mathbf{Q}\cdot\mathbf{u}_{j'}(0)] \right\rangle} = 1 + \left\langle [\mathbf{Q}\cdot\mathbf{u}_j(t)][\mathbf{Q}\cdot\mathbf{u}_{j'}(0)] \right\rangle + \cdots$$

$$\mathbf{u}_j = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}\nu} \left[\hat{u}_{\mathbf{k}\nu} e^{i\mathbf{k}\cdot\mathbf{R}_j} + \hat{u}^+_{\mathbf{k}\nu} e^{-i\mathbf{k}\cdot\mathbf{R}_j} \right] \quad \hat{u}_{\mathbf{k}\nu} \equiv \sqrt{\frac{\hbar}{2M\omega_{\mathbf{k}\nu}}} \mathbf{e}_{\mathbf{k}\nu} \hat{a}_{\mathbf{k}\nu}$$

time dependence $\vec{u}_j(t) = \vec{u}_j e^{-i\omega_{k\nu}t}$ (Heisenberg)

$$\left\langle [\mathbf{Q}\cdot\mathbf{u}_j(t)][\mathbf{Q}\cdot\mathbf{u}_{j'}(0)] \right\rangle = \sum_{\mathbf{k}\nu} \frac{1}{N} \frac{\hbar^2 (\mathbf{e}_{\mathbf{k}\nu}\cdot\mathbf{Q})^2}{2M\hbar\omega_{\mathbf{k}\nu}}$$

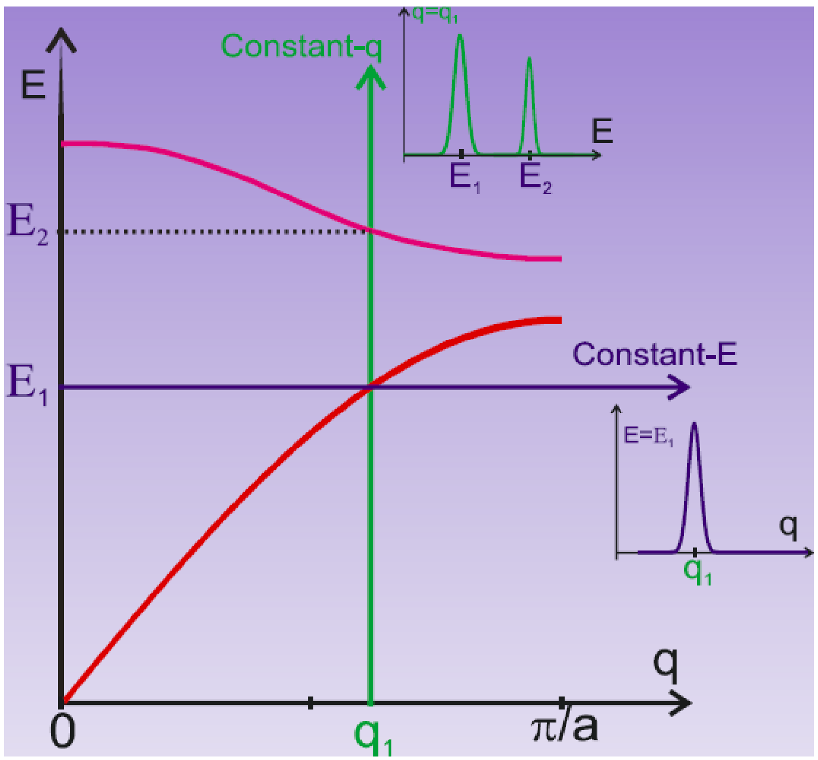
$$\times \left\langle \hat{a}^+_{\mathbf{k}\nu}\hat{a}_{\mathbf{k}\nu} e^{-i\omega_{\mathbf{k}\nu}t} + \hat{a}_{\mathbf{k}\nu}\hat{a}^+_{\mathbf{k}\nu} e^{+i\omega_{\mathbf{k}\nu}t} \right\rangle e^{-i\mathbf{k}\cdot(\mathbf{R}_{j'}-\mathbf{R}_j)}$$

$\langle n \rangle$ $\delta(\omega+\omega_{k\nu})$ $\langle n+1 \rangle$ $\delta(\omega-\omega_{k\nu})$

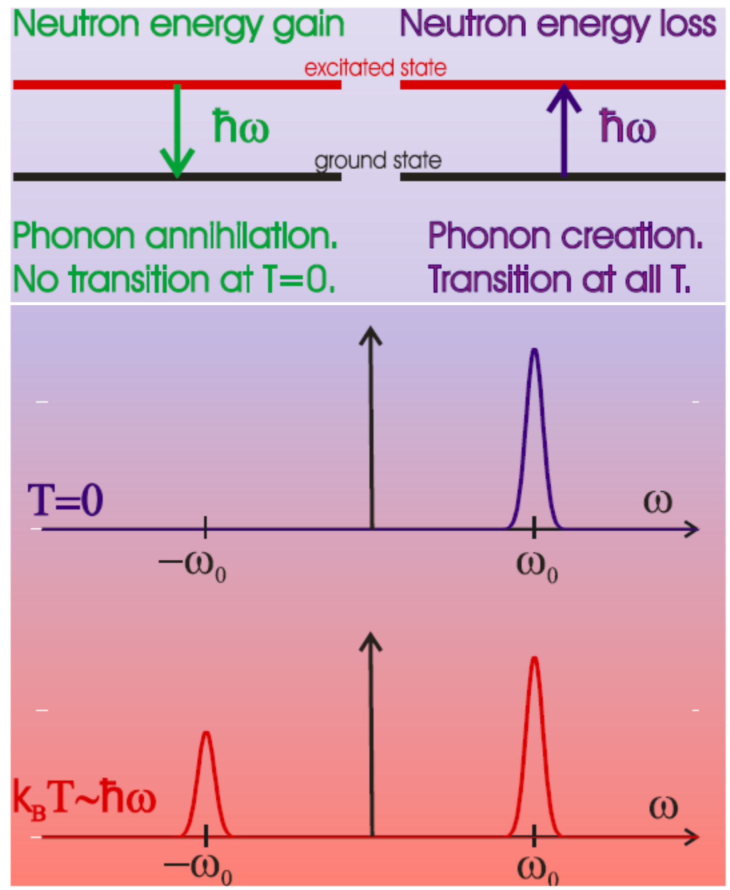
One phonon x-section revisited

$$S(\mathbf{Q},\omega) \propto \frac{(\mathbf{Q}\cdot\mathbf{e}_s)^2}{\omega_{\mathbf{q}s}}\left[\langle n_s+1\rangle\delta(\omega-\omega_{\mathbf{q}s})+\langle n_s\rangle\delta(\omega+\omega_{\mathbf{q}s})\right]$$

Delta function part

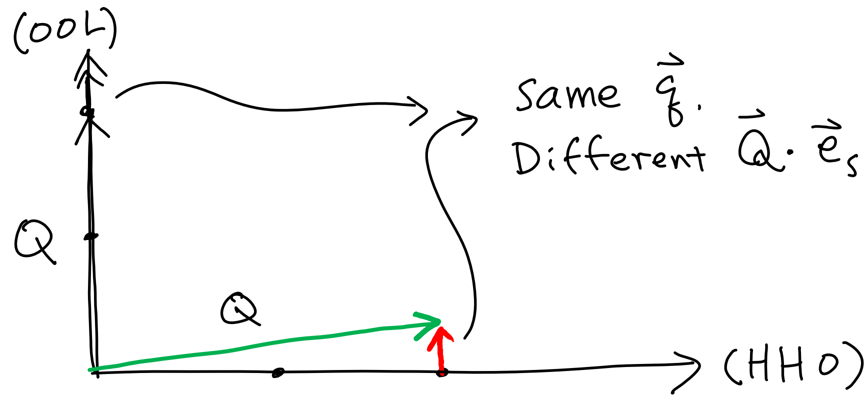


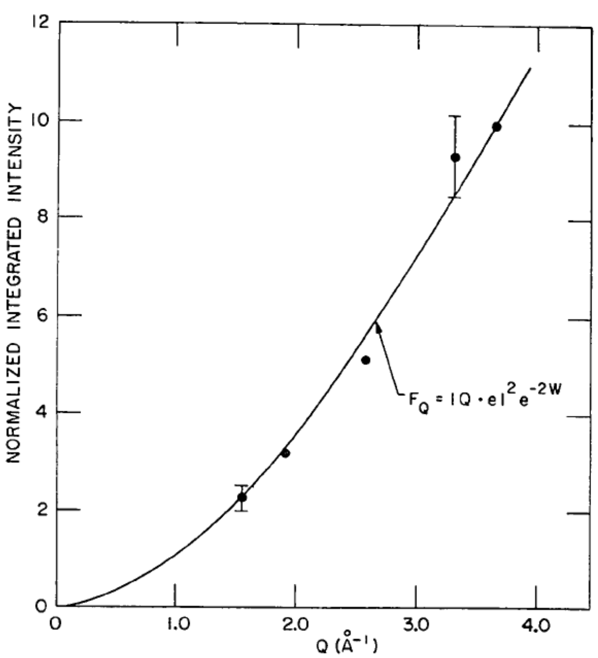
Creation and annihilation



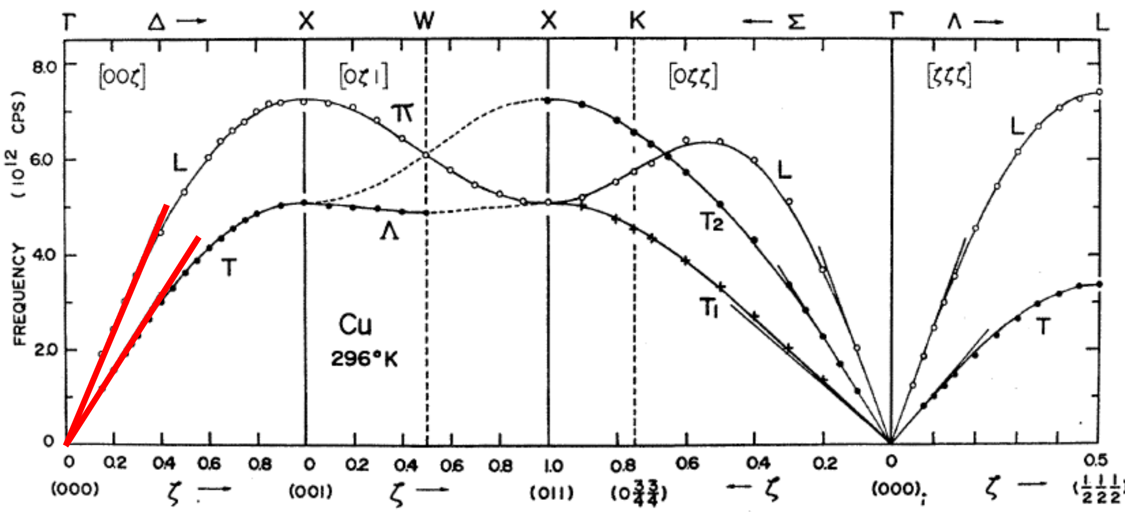
Example of Cu phonon

$$S(\mathbf{Q},\omega) \propto \frac{(\mathbf{Q}\cdot\mathbf{e}_s)^2}{\omega_{\mathbf{q}s}}\left[\langle n_s+1\rangle\delta(\omega-\omega_{\mathbf{q}s})+\langle n_s\rangle\delta(\omega+\omega_{\mathbf{q}s})\right]$$





Chesser and Axe 1973

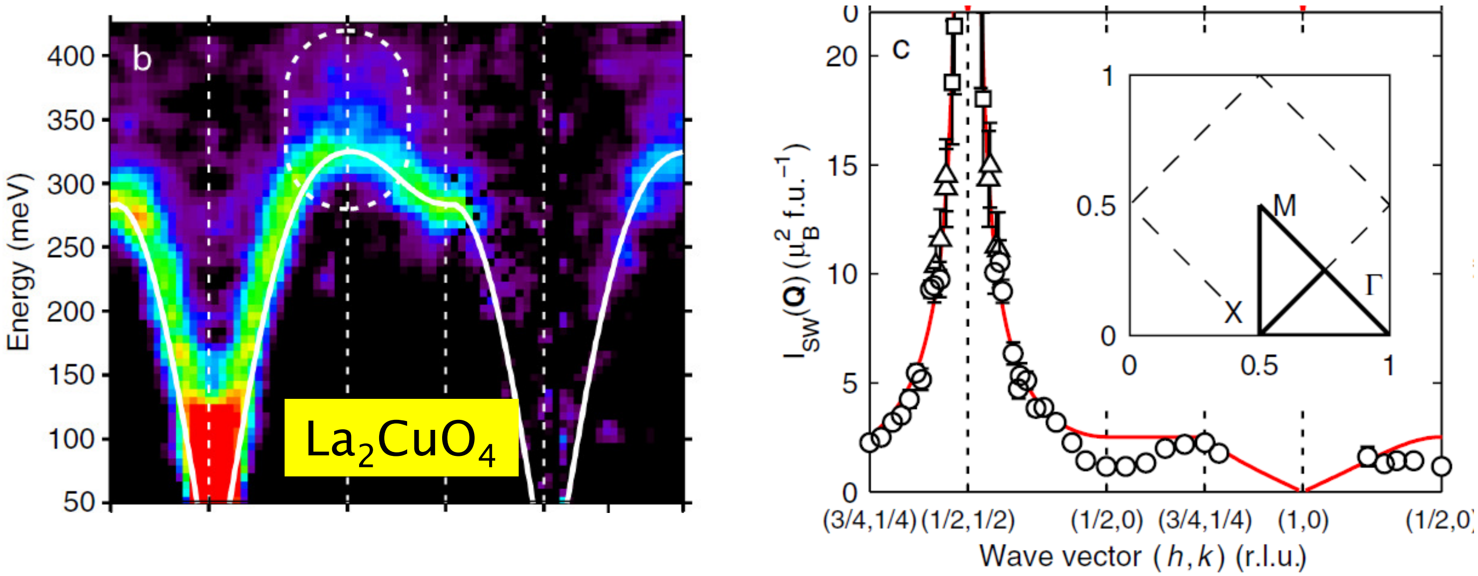


Svensson, Brockhouse, and Rowe 1967

Magnon scattering

$$S(\mathbf{Q},\omega) \propto \sum_{\alpha,\beta}\left(\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta\right)\frac{1}{\omega_{\mathbf{q}s}}\left[\langle n_s + 1\rangle \delta(\omega - \omega_{\mathbf{q}s}) + \langle n_s \rangle \delta(\omega + \omega_{\mathbf{q}s})\right]$$

- Similar to phonon scattering, except for the polarization factor ➔ More by Prof. Broholm
- Magnons ➔ Transverse mode



Headings et al. PRL 105, 247001 (2010)

Damped harmonic oscillator

- In real compounds, delta function broadened due to phonon-phonon and electron-phonon interaction
- Damped harmonic oscillator is used instead

$$\frac{1}{\omega_{\mathbf{q}s}}\delta(\omega \pm \omega_{\mathbf{q}s}) \rightarrow \frac{1}{\pi\omega'_{\mathbf{q}s}}\frac{\Gamma_{\mathbf{q}s}}{(\omega \pm \omega'_{\mathbf{q}s})^2 + \Gamma_{\mathbf{q}s}^2}$$

$$\omega'^2_{\mathbf{q}s} = \omega^2_{\mathbf{q}s} - \Gamma^2_{\mathbf{q}s}$$

- What about overdamping? $\omega_{\mathbf{q}s} < \Gamma_{\mathbf{q}s}$
  - Diffusion ➔ Quasielastic scattering
  - Critical scattering

Conclusions

- Studying excitations are essential for understanding materials properties
- INS can tell us about
  - Phonons
  - Magnons
  - Diffusive excitations
  - Molecular level excitations
- INS from phonons
  - Energy loss vs. energy gain
  - Use of Q
    - phonon polarization vector
    - High Q Brillouin zone