Introduction to neutron spectroscopy

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Neutron scattering cross-section

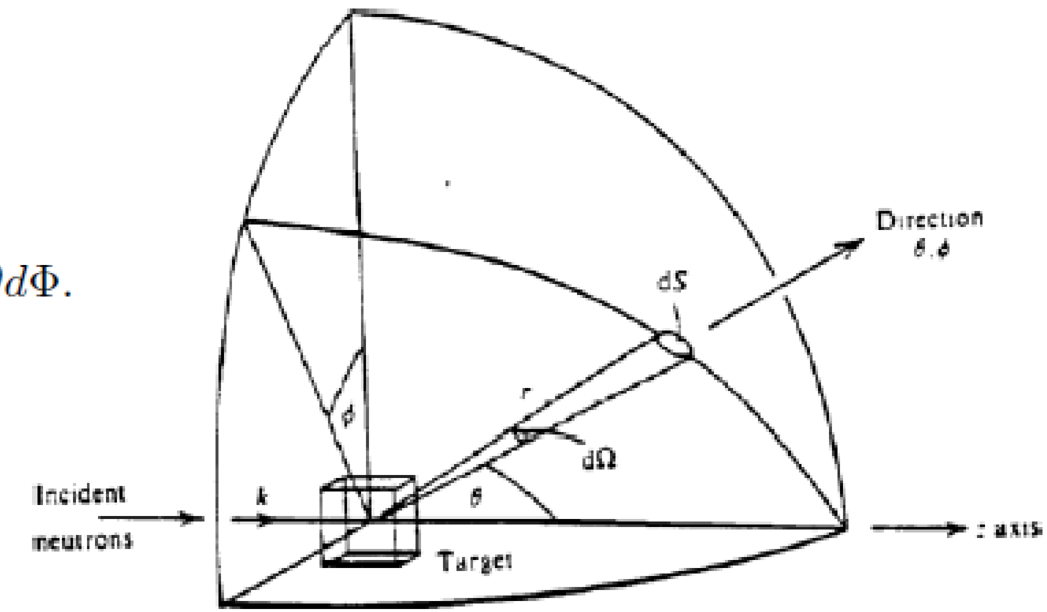
Theory

Partial differential cross-section

Definition:

The partial differential cross-section $\frac{d\sigma}{d\Omega dE'}$ gives the fraction of neutrons that can be scattered by a target in a solid angle $d\Omega$ with a final energy between E' and $E' + dE'$. $\frac{d\sigma}{d\Omega dE'}$ has the dimension of an area.

$$d\Omega = sin(\theta)d\theta d\Phi.$$



Partial differential cross-section

Neutron:

no charge	spin ½	plane wave	energy	state
		$\psi_k(\vec{r}) = \frac{1}{\sqrt{V}} e^{i\vec{k}\vec{r}}$	$E_k = \frac{\hbar^2 k^2}{2M}$	$\vert k, \sigma >$

Target:

energy	state
E_λ	$\vert\lambda >$

Scattering:

initial state $|k\sigma\lambda >$ → final state $|k'\sigma'\lambda' >$

Kinematic constraints

$$\begin{aligned} E_\lambda + E_k &= E_{\lambda'} + E_{k'} \\ \hbar\vec{Q} &= \hbar(\vec{k} - \vec{k}') \end{aligned}$$

Partial differential cross-section

Probability for the incident neutron to be in the spin state |σ>

Density of accessible states k'

$$D = \frac{V}{(2\pi)^3} d\vec{k'}$$
$$= \frac{V}{(2\pi)^3} k' \frac{M}{\hbar^2} dE_{k'} d\Omega$$

$$d\sigma = [\frac{1}{F}] . \sum_{\sigma,\sigma'} p_\sigma \sum_{\lambda,\lambda'} p_\lambda [W].[D]$$

Neutron flux

$$F = \frac{1}{V}\frac{\hbar k}{M}$$

Probability for the target to be in the initial state |λ>

$$p_\lambda = \frac{e^{-\beta E_\lambda}}{\sum_\lambda e^{-\beta E_\lambda}}$$

Partial differential cross-section

$$d\sigma = [\frac{1}{F}].\sum_{\sigma,\sigma'} p_\sigma \sum_{\lambda,\lambda'} p_\lambda [W].[D]$$

$\Box$ W corresponds to the probability of a transition from $|k\sigma\lambda>$ to $|k'\sigma'\lambda'>$
This probolility is given by the Fermi's golden rule:

$$W = \frac{2\pi}{\hbar}| < k'\sigma'\lambda'|\hat{V}(\vec{r})|k\sigma\lambda > |^2 \delta(E_\lambda + E_k - E_{\lambda'} - E_{k'})$$

Scattering potential

Partial differential scattering cross-section

$$\frac{d\sigma}{d\Omega dE'} = \frac{k}{k'}(V\frac{M}{2\pi\hbar^2})^2 \sum_{\sigma,\sigma'} p_\sigma \sum_{\lambda,\lambda'} p_\lambda | < k'\sigma'\lambda'|\hat{V}(\vec{r})|k\sigma\lambda > |^2 \delta(\hbar\omega + E_\lambda - E_{\lambda'})$$

Nuclear neutron scattering

Theory

Nuclear scattering: scattering by a single nucleus

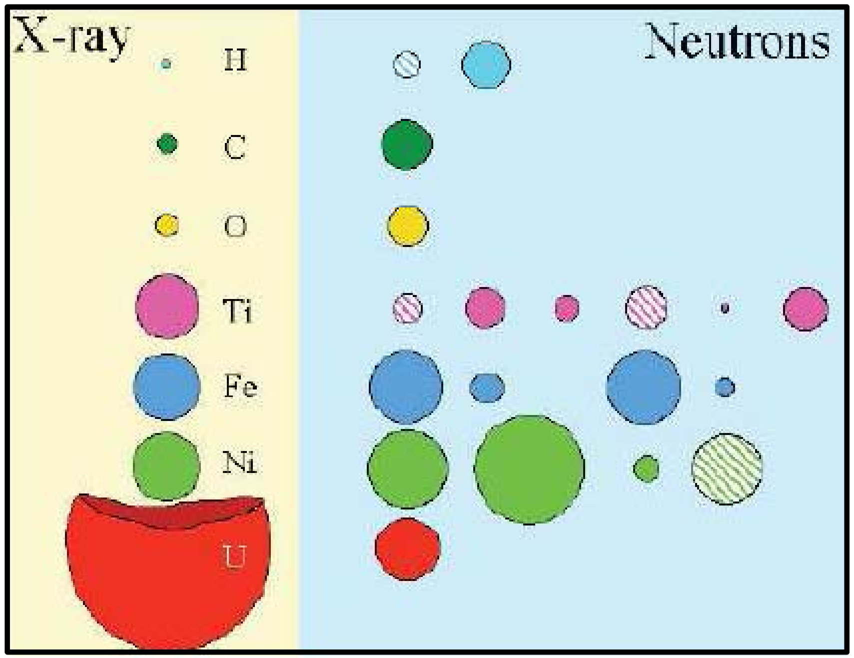
$$\hat{V}_n(\vec{r}) = \frac{2\pi\hbar^2}{M} b\delta(\vec{r} - \vec{R}).$$

$$\Downarrow$$

b= scattering length

- *positive or negative*
- *depend on the isotope*

Scattering strength



Nuclear scattering: scattering by a single nucleus

• **remark (1):**
For a full description of the nuclear scattering, one should also include the interaction with the nuclear spin $\hat{I}$ of the nucleus.

$$b = b_0 + \frac{1}{2} b_n \hat{I} \hat{\sigma} \tag{2.7}$$

One usually calls b^+ and b^- the two values of b depending on the neutron spin state $|+>$ or $|->$. Since $J = I + \frac{1}{2}$ is a good quantum number, one obtains:

$$b^+ = b_0 + \frac{1}{2} b_n I \tag{2.8}$$

$$b^- = b_0 + \frac{1}{2} b_n (I+1) \tag{2.9}$$

• **remark (2):**
There are a few nuclei that can also capture the neutron: the neutron forms a bound state with the nucleus. To account for this absorption, one introduce an imaginary part for b, called b", related to σ_a the absorption scattering cross-section:

$$b" = \frac{k_0}{4\pi} \sigma_a \tag{2.10}$$

• **remark (3):**
For sake of simplicity, we will not consider neither the magnetic interaction of a neutron with a nucleus or the obsortion effect in th rest of this chapter.

Nuclear scattering: scattering by a single nucleus

Non magnetic scattering :
$$\sum_{\sigma,\sigma'} p_\sigma < \sigma|\sigma' >< \sigma'|\sigma >= \sum_{\sigma,\sigma'} p_\sigma \delta_{\sigma,\sigma'} = 1$$

Evaluation of the term : $< k'|\hat{V}_n(\vec{r})|k >$

$$V\frac{M}{2\pi\hbar^2} < k'|\hat{V}_n(\vec{r})|k >= \int d\vec{r}\psi^\dagger_{k'}(\vec{r})\hat{V}_n(\vec{r})\psi_k(\vec{r}) = be^{i\vec{Q}\vec{R}}$$

⇨ $$\frac{d\sigma}{d\Omega dE'} = \frac{k}{k'}\frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} dte^{i\omega t}\sum_{\lambda,\lambda'} p_\lambda < \lambda|b^\star e^{-i\vec{Q}\vec{R}(0)}|\lambda' >< \lambda'|be^{i\vec{Q}\vec{R}(t)}|\lambda >$$

Nuclear scattering: scattering by a single nucleus

Properties of the δ-function: $\delta(x) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} dt e^{ixt}$ $\qquad \delta(ax) = \frac{1}{|a|}\delta(x)$

Time dependent operator: $\hat{A}(t) = e^{-i\frac{Ht}{\hbar}}\hat{A}(t=0)e^{i\frac{Ht}{\hbar}}$

Projector: $\sum_{\lambda} |\lambda' >< \lambda'| = 1$

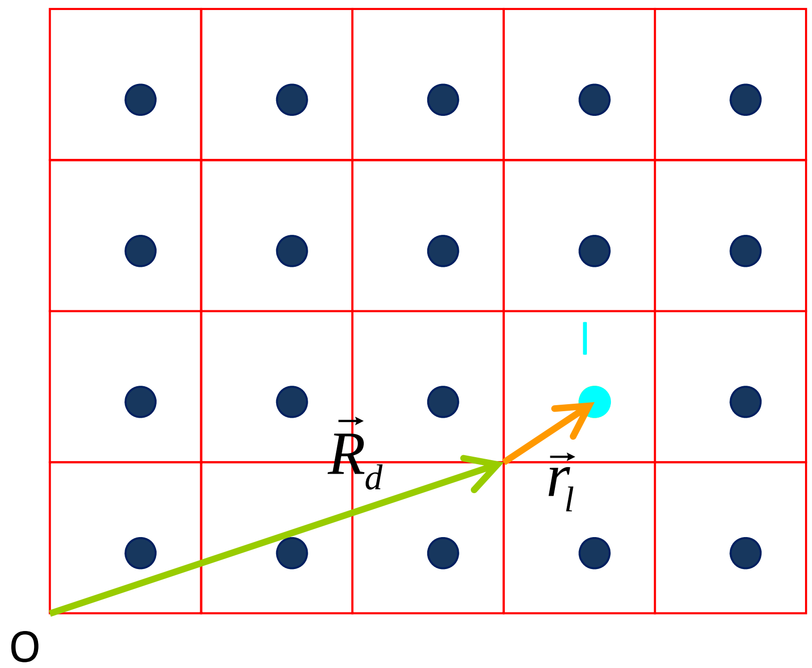
Thermodynamic average: $\sum_{\lambda} p_{\lambda} < \lambda|...|\lambda > = < ... >_T$

$$\frac{d\sigma}{d\Omega dE'} = \frac{k}{k'}\bar{b}^2 \frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} dt e^{i\omega t} < e^{-i\vec{Q}\vec{R}(0)} e^{i\vec{Q}\vec{R}(t)} >_T$$

Nuclear scattering: crystal

A crystal is made of a periodic arrangement of nuclei. The buiding block of a crystal is the unit cell which contains a specific arrangement of n nuclei. For a crystal with a periodic repetition of N unit cells, the total volume $V = Nv_0$, where v_0 is the volum of the unit cell. Each type of nuclei of the unit cell is identified by the index l. Its scattering length is b_l and its mass is m_l.

The location of a nucleus in the crystal is given by a sum of three terms: $\vec{R}_d + \vec{r}_l + \vec{u}_{d,l}(t)$.
- $\vec{R}_d$ is a vector of the Bravais lattice and indicates the unit cell the nucleus belongs to.
- $\vec{r}_l$ is its mean position in the unit cell.
- $\vec{u}_{d,l}(t)$ is its displacement with respect to its mean position.



Nuclear scattering: crystal

Scattering potential :

$$\hat{V}_n(\vec{r}) = \frac{2\pi\hbar^2}{M}\sum_{d=1}^{N}\sum_{l=1}^{n} b_l\delta(\vec{r}-\vec{R}_d-\vec{r}_l-\vec{u}_{d,l})$$

$$\frac{d\sigma}{d\Omega dE'} = \frac{k}{k'}\sum_{d,d'} e^{-i\vec{Q}(\vec{R}_d-\vec{R}'_d)}\sum_{l,l'} b_l b_{l'} e^{-i\vec{Q}(\vec{r}_l-\vec{r}'_l)}\frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} dt e^{i\omega t} < e^{-i\vec{Q}\vec{u}_{d,l}(0)} e^{i\vec{Q}\vec{u}_{d',l'}(t)} >_T$$

periodic arrangment of the lattice

Organisation of atoms within the unit cell

???

Nuclear scattering: crystal

using the so-called Bloch identity

$$< e^{-i\vec{Q}\vec{u}_{d,l}(0)} e^{i\vec{Q}\vec{u}_{d',l'}(t)} >_T = e^{-W_l} e^{-W_{l'}} e^{<\vec{Q}\vec{u}_{d,l}(0).\vec{Q}\vec{u}_{d',l'}(t)>_T}$$

Debye-Waller factor:

$$W_l = \frac{1}{2} < [\vec{Q}\vec{u}_{d,l}]^2 >_T$$

small displacement with respect to $|Q|^{-1}$

$$e^{<\vec{Q}\vec{u}_{d,l}(0).\vec{Q}\vec{u}_{d',l'}(t)>_T} \sim 1 + < \vec{Q}\vec{u}_{d,l}(0).\vec{Q}\vec{u}_{d',l'}(t) >_T$$

Elastic term

Inelastic terms

$\vec{u}_{d,l}(t)$ is induced by the collective vibrations of nuclei, the phonon modes. One uses the label s to identify a given phononic mode and $\hat{e}^s_{\vec{q}}$ describes the polarisation of the mode with respect to the propagation wave vector $\vec{q}$. $\hbar\omega^s_{\vec{q}}$ is the phonon energy .
Notice that $\vec{q}$ belongs to the first Brillouin Zone.

Nuclear scattering: crystal

$\vec{Q}$ is expressed as a combination of a vector $\vec{\tau}$ of the reciprocal lattice and $\vec{q}$ of the first Brillouin Zone:

$$\vec{Q} = \vec{\tau} + \vec{q}.$$

Notice that $\vec{\tau}\vec{R}_d = 2\pi m$ with m integer and $e^{i\vec{\tau}\vec{R}_d} = 1$.
Furthermore, $\sum_{d,d'} e^{i\vec{Q}(\vec{R}_d - \vec{R}_{d'})} = N\frac{(2\pi)^3}{v_0}\sum_{\vec{\tau}}\delta(\vec{Q}-\vec{\tau})$

The coherent partial differential cross-section has an elastic term and an inelastic term:

$$\boxed{\frac{d\sigma}{d\Omega dE'} = \left(\frac{d\sigma}{d\Omega dE'}\right)_{elas} + \left(\frac{d\sigma}{d\Omega dE'}\right)_{inelas}}$$

Nuclear scattering: Elastic scattering cross-section

$$\left(\frac{d\sigma}{d\Omega dE'}\right)_{elas} = N\frac{(2\pi)^3}{v_0}\sum_{\vec{\tau}}\delta(\vec{Q}-\vec{\tau})\left|\sum_l b_l e^{-W_l} e^{-i\vec{Q}\vec{r}_l}\right|^2 \delta(\omega)$$

$k = k'$

Elastic structure factor
⟹ crystal structure

Debye-Waller factor

Bragg reflections in reciprocal space

Nuclear scattering: Inelastic scattering cross-section

$n_B(\omega) = \frac{1}{e^{\beta\hbar\omega}-1}$ the Bose-Einstein distribution

Dynamical structure factor
phonons

$$\left(\frac{d\sigma}{d\Omega dE'}\right)_{inelas} = \frac{k'}{k} N \frac{(2\pi)^3}{v_0} \sum_{\vec{\tau}} \delta(\vec{Q}-\vec{\tau}-\vec{q}) \left| \sum_l b_l e^{-W_l} e^{-i\vec{Q}\vec{r}_l} \left(\frac{\hbar}{2m_l\omega_{\vec{q}}^s}\right)^{\frac{1}{2}} \hat{e}_{\vec{q}}^s \vec{Q} \right|^2$$

$$\times \left((1+n_B(\omega_{\vec{q}}^s))\delta(\omega-\omega_{\vec{q}}^s) + n_B(\omega_{\vec{q}}^s)\delta(\omega+\omega_{\vec{q}}^s) \right)$$

creation

anihilation

polarization

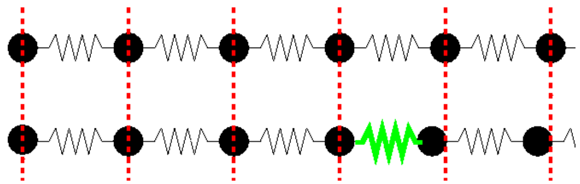
A specific choice of $\vec{Q}$ provides detailed information on the polarization vector $\hat{e}_{\vec{q}}^s$.

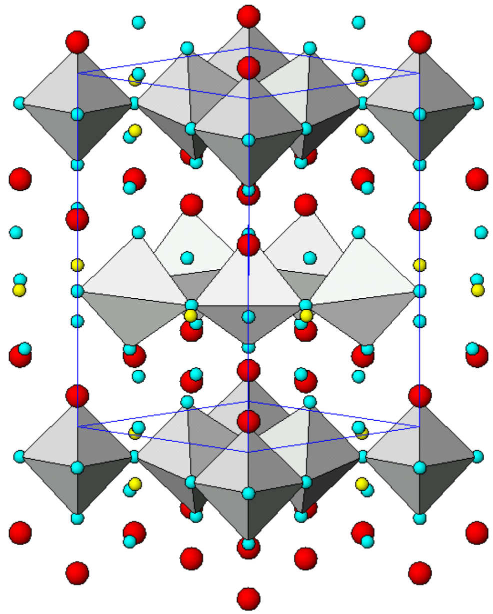
Lattice vibration(1)

Atoms oscillating in harmonic potentials

$$H = \frac{1}{2} M_\ell \left(\frac{d}{dt} u \right)^2 + \frac{1}{2} \Gamma u^2$$

$$F = -\mathrm{grad}\ V = -\Gamma u$$





Sum of all forces on a given atom

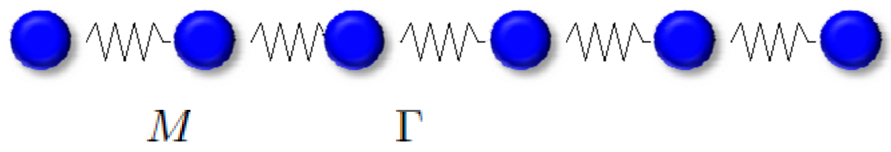
$$F_{m,\ell,\alpha} = - \sum_{n,\ell',\beta} \Gamma^{\alpha,\beta}_{m,\ell,n,\ell'}\ u_{n,\ell',\beta}$$

Equation of motion

$$M_\ell \frac{d^2}{dt^2} u_{m,\ell,\alpha} = F_{m,\ell,\alpha} = - \sum_{n,\ell',\beta} \Gamma^{\alpha,\beta}_{m,\ell,n,\ell'}\ u_{n,\ell',\beta}$$

Lattice dynamics (2)

Mono-atomic chain



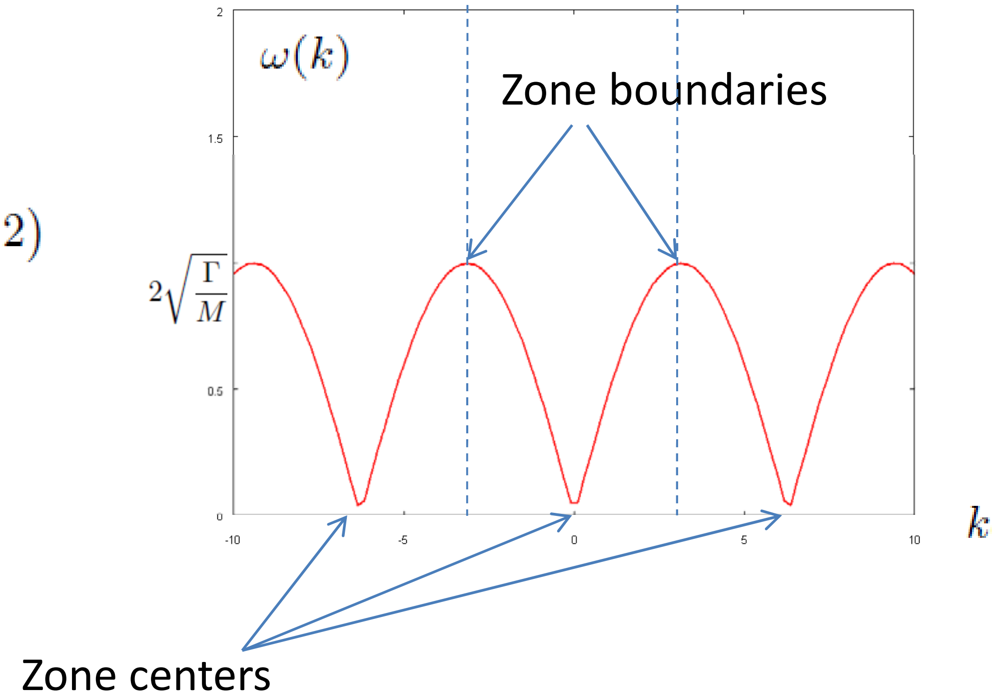
$$M\frac{d^2}{dt^2}u_m = \Gamma\ (u_{m+1} + u_{m-1} - 2u_m)$$

Fourier transform

$$-M\omega^2 = \Gamma\ (e^{ika} + e^{-ika} - 2)$$

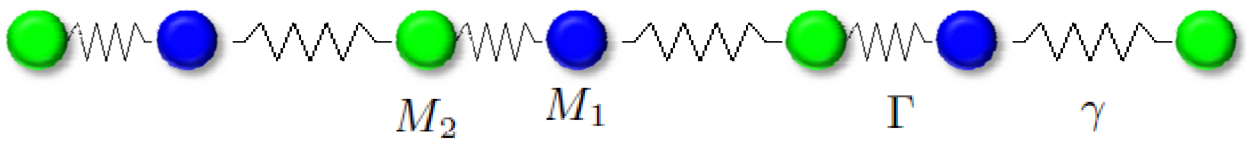
$$\omega(k) = 2\sqrt{\frac{\Gamma}{M}}\sin\frac{ka}{2}$$

Dispersion relation



Lattice dynamics (3)

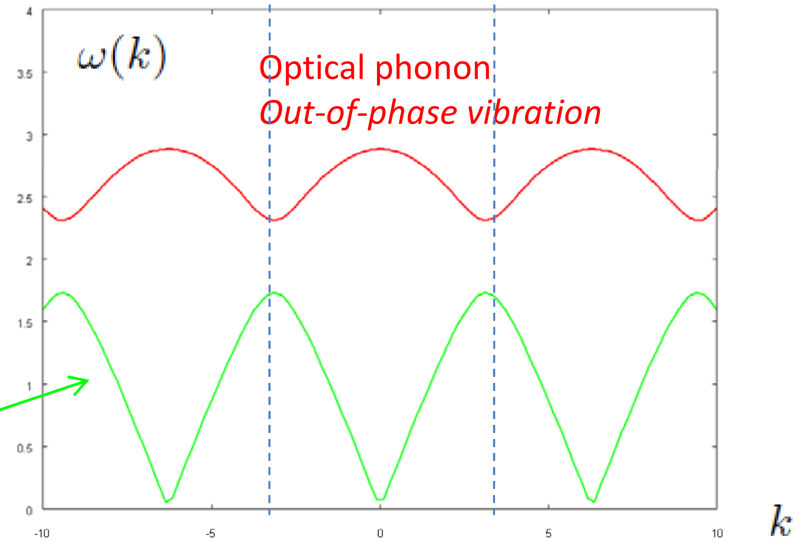
Di-atomic chain



$$
\begin{aligned}
M_1 \frac{d^2}{dt^2} u_{m,1} &= \Gamma \ (u_{m,2} - u_{m,1}) + \gamma \, (u_{m-1,2} - u_{m,1}) \\
M_2 \frac{d^2}{dt^2} u_{m,2} &= \Gamma \ (u_{m,1} - u_{m,2}) + \gamma \, (u_{m+1,1} - u_{m,2})
\end{aligned}
$$

Fourier transform: 2 sets of equations

$$
\begin{pmatrix} \omega^2 - \frac{\Gamma+\gamma}{M_1} & \frac{\Gamma+\gamma e^{-ika}}{\sqrt{M_1 M_2}} \\ \frac{\Gamma+\gamma e^{ika}}{\sqrt{M_1 M_2}} & \omega^2 - \frac{\Gamma+\gamma}{M_2} \end{pmatrix} \begin{pmatrix} \sqrt{M_1} u_1 \\ \sqrt{M_2} u_2 \end{pmatrix} = 0
$$



Lattice dynamics (3)

General case

$$M_\ell \frac{d^2}{dt^2} u_{m,\ell,\alpha} = F_{m,\ell,\alpha} = -\sum_{n,\ell',\beta} \Gamma^{\alpha,\beta}_{m,\ell,n,\ell'} \, u_{n,\ell',\beta}$$

Normal coordinates

$$q_{k,\ell,\alpha} = \sqrt{M_\ell} \, u_{k,\ell,\alpha}$$

Fourier transform:

$$\frac{d^2}{dt^2} q_{k,\ell,\alpha} = -\sum_{n,\ell',\beta} \frac{\Gamma^{\alpha,\beta}_{k,\ell,\ell'}}{\sqrt{M_\ell M_{\ell'}}} \, q_{k,\ell',\beta}$$

$D^{\alpha,\beta}_{k,\ell,\ell'}$ Dynamical matrix of dimension $L \times d$

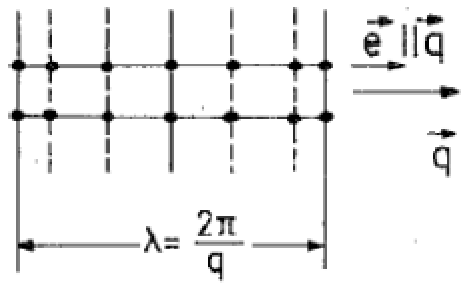
$$\omega^2 q_{k,\ell,\alpha} = D^{\alpha,\beta}_{k,\ell,\ell'} \, q_{k,\ell',\beta}$$

Eigenmodes = phonons = collective vibration modes

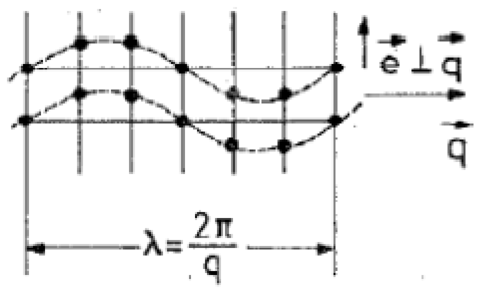
$\omega^2_{k,s}$ o square root of the phonon frequency

$e^{(s)}_k$ o phonon polarization

Longitundinal modes



Transverse modes



Magnetic neutron scattering

Theory

Magnetic scattering potentials

Scattering potential: $\hat{V} = -\hat{\mu}.\vec{H}$

<u>Neutron:</u> magnetic moment operator

$$\hat{\mu} = \gamma \mu_N \hat{\sigma}$$

<u>Target</u>: distribution of internal magnetic fields

➡ (1) Spins of unpaired electron
(2) Electronic orbital moments
(3) Nuclear spins

spin-only scattering: Dipolar interaction with electronic spins

$$\vec{H} = c\vec{u}rl \left(\frac{2\mu_B \hat{s} \times \vec{R}}{R^3} \right)$$

unpolarized neutron beam: $\sum_\sigma p_\sigma < \sigma|\sigma_\alpha \sigma_\beta|\sigma >= \delta_{\alpha,\beta}$

Partial differential cross-section

prefactor

$$r_0 = 4\pi(\frac{M}{2\pi\hbar^2})2\gamma\mu_N\mu_B$$
$$= \frac{\gamma e^2}{m_e c^2}$$

Owing to the dipolar Interaction one can observe only the spin components perp. to Q

$$\frac{d\sigma}{d\Omega dE'} = \frac{k'}{k} r_0^2 \sum_{\alpha,\beta} \left(\delta_{\alpha,\beta} - \frac{Q_\alpha Q_\beta}{Q^2}\right)$$
$$\times \sum_{l',l'} F_l(\vec{Q}) F_{l'}(\vec{Q}) e^{i\vec{Q}(\vec{R}_l - \vec{R}_{l'})}$$
$$\times \sum_{\lambda,\lambda'} p_\lambda < \lambda | \hat{s}_{l,d,\alpha} | \lambda' >< \lambda' | \hat{s}_{l',d',\beta} | \lambda > \delta(\hbar\omega + E_\lambda - E_{\lambda'})$$

Magnetic form factor = Fourier transform of the spin Distribution on each atom

Magnetic structure factor

For a single type of magnetic atom

$$\frac{d\sigma}{d\Omega dE'} = \frac{k'}{k} F(\vec{Q})^2 \sum_{\alpha,\beta} \left(\delta_{\alpha,\beta} - \frac{Q_\alpha Q_\beta}{Q^2} \right) S_{\alpha,\beta}(\vec{q}, \omega)$$

$$S_{\alpha,\beta}(\vec{Q}, \omega) = \frac{1}{N} \frac{1}{2\pi\hbar} \int_{-\infty}^{+\infty} dt e^{i\omega t} \sum_{l,l'} e^{i\vec{Q}(\vec{R}_l - \vec{R}_{l'})} < S_{\alpha,l}(0) S_{\beta,l'}(t) >_T$$

Magnetic structure factor = Fourier transform in space and time of spin-spin correlation function

Fluctuation-dissipation theorem & detailed balance factor

Detailed balance factor $[1 + n_B(\omega)] = \frac{1}{1 - e^{-\beta\hbar\omega}}$

$$S_{\alpha,\beta}(\vec{Q},\omega)|_{\omega\neq 0} = \frac{1}{\pi}\frac{1}{1 - e^{-\beta\hbar\omega}} Im\chi_{\alpha,\beta}(\vec{Q},\omega)$$

Imaginary part of the dynamical magnetic susceptibility

$S_{\alpha,\beta}(\vec{Q},\omega)$ is the Fourier transform in space and time of the spin-spin correlation function. Likewise the dynamical magnetic structure factor is proportional to the imaginary part of the dynamical spin susceptibility.

Ordered magnetic moment & spin waves

$$\frac{d\sigma}{d\Omega dE'} = \left(\frac{d\sigma}{d\Omega dE'}\right)_{elas} + \left(\frac{d\sigma}{d\Omega dE'}\right)_{inelas}$$

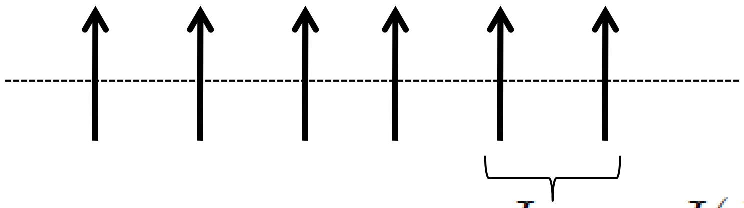
$$\left(\frac{d\sigma}{d\Omega dE'}\right)_{elas} \propto < S_\perp(Q)^2 >_T \delta(\omega)$$

$$\left(\frac{d\sigma}{d\Omega dE'}\right)_{inelas} \propto \sum_s A_s[(1 + n_B(\omega_{q,s}))\delta(\omega - \omega_{q,s}) + (n_B(\omega_{q,s}))\delta(\omega + \omega_{q,s})]$$

In the case of a magnetic order, the elastic structure factor describes the ordered magnetic pattern and is proportional to the square of the ordered magnetic moment. As in the case of phonons, the dynamical structure factor is related to the creation or the anihilation of (collective) magnetic excitations, i.e magnons of energy $\hbar\omega_{\vec{q}}$ (in general): $Im\chi(\vec{Q},\omega) \propto [\delta(\omega-\omega_{\vec{q}})-\delta(\omega+\omega_{\vec{q}})]$. Note that, in agreement with spin wave theory, it has to be proportional the ordered magnetic moment, whereas the static structure factor is proportional to the square of the ordered magnetic moment.

Spin wave – Magnons (1)

Ferromagnetic chain:



$\vec{S}_i = S\hat{z}$

$J_{i,j} = J(\vec{R}_i - \vec{R}_j)$

Hamiltonian: $H = -\sum_{i,j} J_{i,j}\vec{S}_i\vec{S}_j$

Spin fluctuations: $\vec{S}_i \simeq S\hat{z} + \delta S_x\hat{x} + \delta S_y\hat{y}$

Eqn of motion: $\dfrac{d\vec{S}_i}{dt} = \vec{S}_i \times g\mu_B\vec{h}_o$

Mean field approx. $g\mu_B\vec{h}_o = 2\sum_{i,j} J_{i,j}\vec{S}_i$

Eqn of motion: $\dfrac{d\delta\vec{S}_i}{dt} = 2S\sum_{j} J_{i,j}(\delta\vec{S}_i - \delta\vec{S}_j) \times \hat{z}$

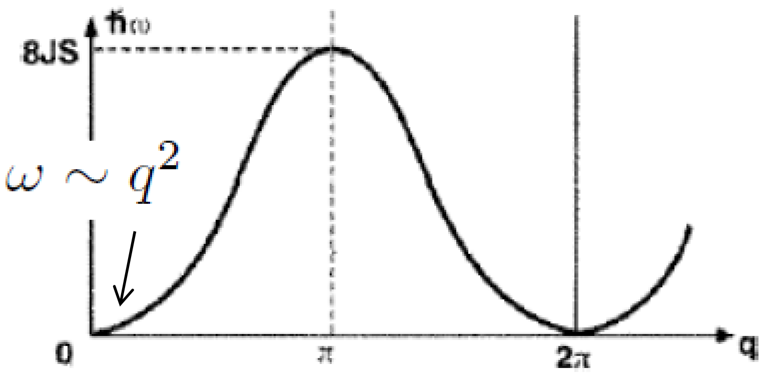
Spin wave – Magnons (2)

Fourier components: $\vec{S}(q) = \sum_l e^{-i\vec{q}\vec{R}_l}$ and $J(q)$

2coupled eqns

$$\frac{d\delta S_x(q)}{dt} = +2S[J(0) - J(q)]\delta S_y(q)$$
$$\frac{d\delta S_y(q)}{dt} = -2S[J(0) - J(q)]\delta S_x(q)$$

Fourier components: $S^-(q) = S_x(q) - iS_y(q) = Ns(q)e^{i\omega t}$

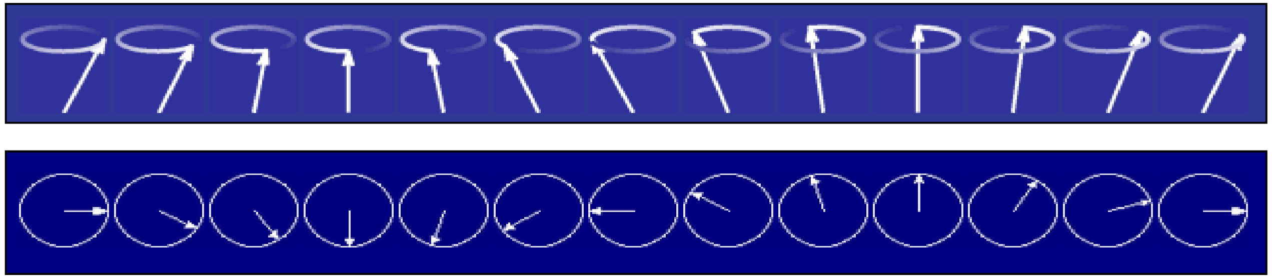


$$\boxed{\omega_q = 2S[J(0) - J(q)]}$$

$$J(q) = 2Jcosq_a$$

Spin wave – Magnons (3)

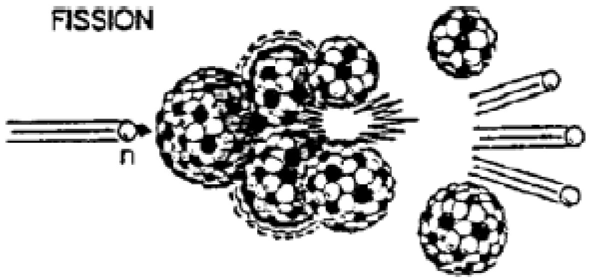
Spin wave: $\delta\vec{S}_i = s(q)[cos(\vec{q}\vec{R}_i + \omega t)\hat{x} + sin(\vec{q}\vec{R}_i + \omega t)\hat{y}]$

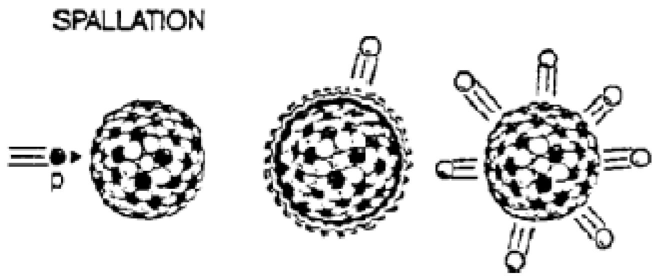


Neutron scattering technique

Theory versus Reality

Neutron sources





-(1) a Fission reactor: a thermal neutron is absorbed by a ^{235}U nucleus, which splits into fission fragments and evaporates a few neutrons with energy of 1 to 2 MeV.

-(2) spallation: A high energy proton (~1GeV) chops pieces of a heavy nucleus. Twenty to forty neutrons are evaporated with energies of typically a few MeV

Neutron energy and wave length

Neutrons then slow down by loosing energy through inelastic collisions with light atoms (H,D,Be).

Characteristic energy (meV).

0.1 - 10 meV : cold neutrons (moderator: liquid H_2 at 20 K)

10 - 100 meV : thermal neutrons

100 - 500 meV : hot neutrons (moderator: graphite at 2000 K)

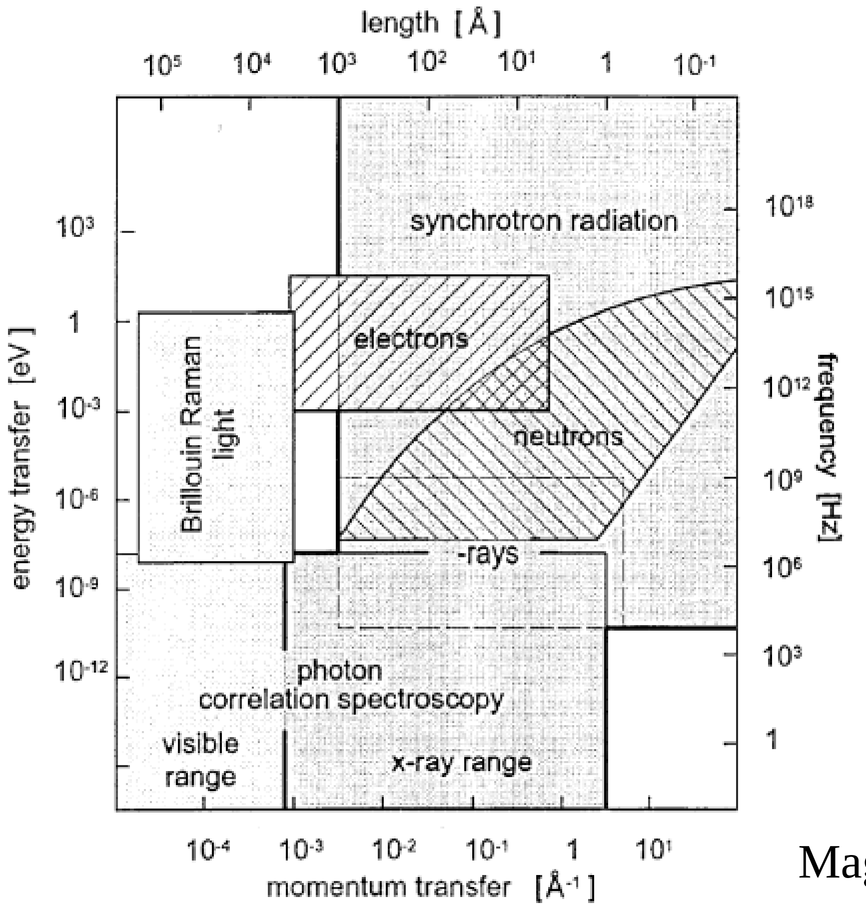
> 500 meV : epithermal neutrons

A neutron of 25 meV $\Longrightarrow$ wave length of ~1.48 °A.
the same order of magnitude as the typical inter-atomic distance in a solid.

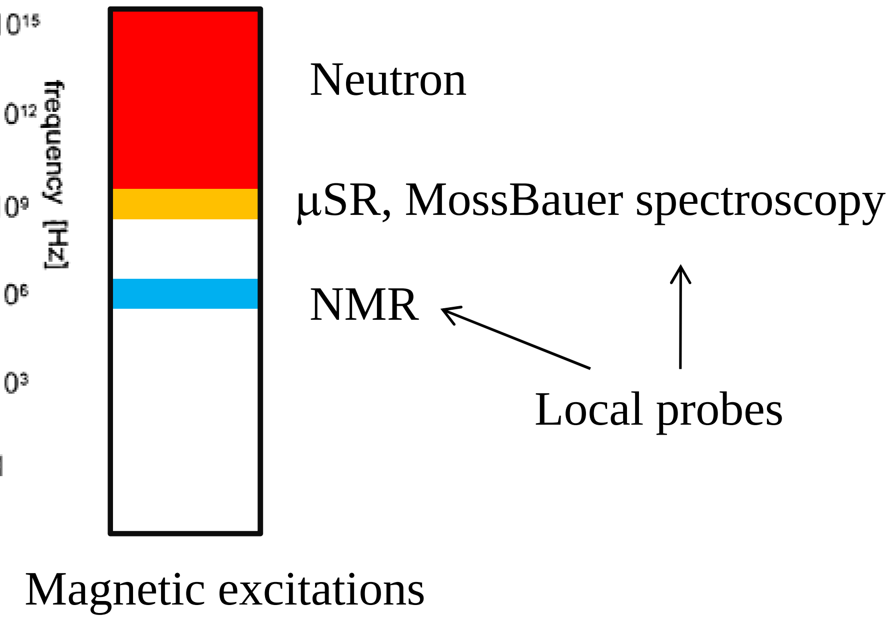
Neutron energy $\Longrightarrow$ same order of magnitude as the characteristic excitations in a solid such as the phonons or the magnons.

Neutron energy and wave length

Neutron spectroscopy can be used to probe the nuclear and magnetic structures of a sample and the related nuclear and magnetic excitations. This is a <u>bulk</u> and <u>non destructive</u> measurement.



Neutron scatering vs. Other inelastic probes

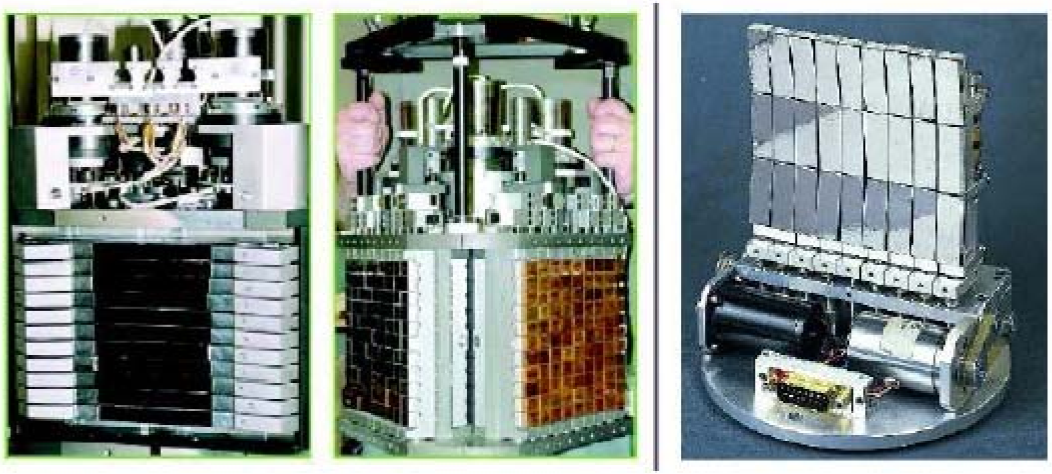


Monochromatic neutron beam

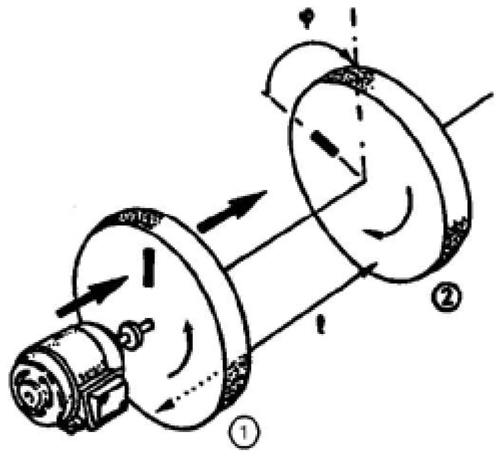
Monochromator:

Bragg scattering on a
single crysral

$$2dsin(\theta) = \lambda$$



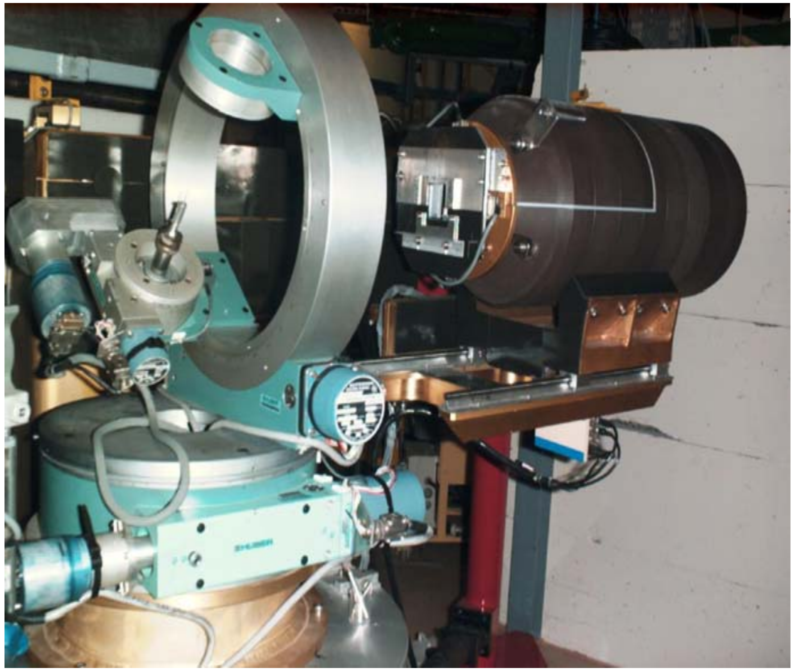
Chopper :



Diffraction measurements : integration on the final energy

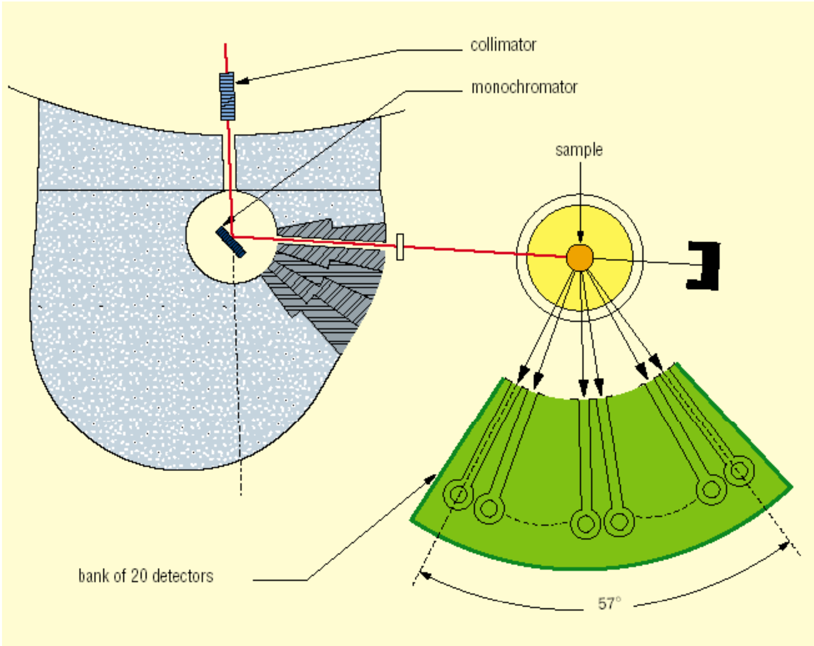
single crystals
4-circle diffractometers

$$\vec{Q}$$



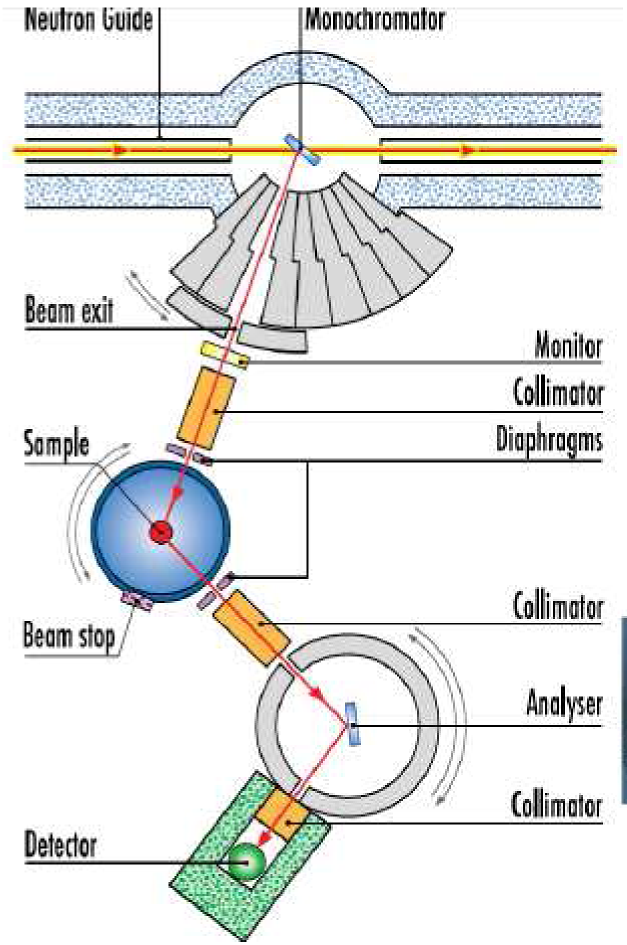
Powder sample
Powder diffractometer

$$\left|\vec{Q}\right|$$

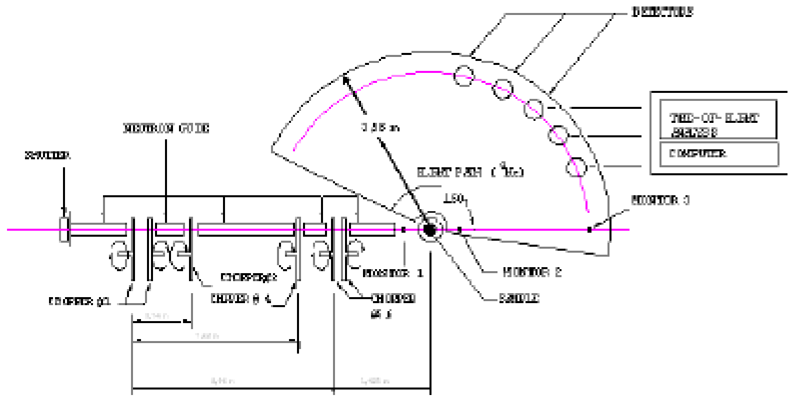


Inelastic Measurements: analysis of the final neutron energy

Single crystal
Triple-axis spectrometer
(TAS)



Powder samples
Time of flight spectrometer
(TOF)



The final energy is given: ½ Mv^2

$$v = D/\tau.$$

D is the fixed distance between the sample and the detector (*the flight path*) and τ the time of flight of the neutron.

Instrumental resolution

Measured intensity proportional to $S(\vec{Q},\omega) \otimes R(\vec{Q},\omega)$

The Fourier transform of the theoretical structure factor convoluted by the instrumental resolution function

$R(\vec{Q},\omega)$ ⇨ Gaussian function (4 dimensions)
Resolution ellipsoid

Simplified description (Cooper, Nathans,1967…)
The resolution function reads : $R_0 \exp(-X^t \mathbf{A} X)$
X stands for a 4D vector : $(Q-Q_0, w-w_0)$

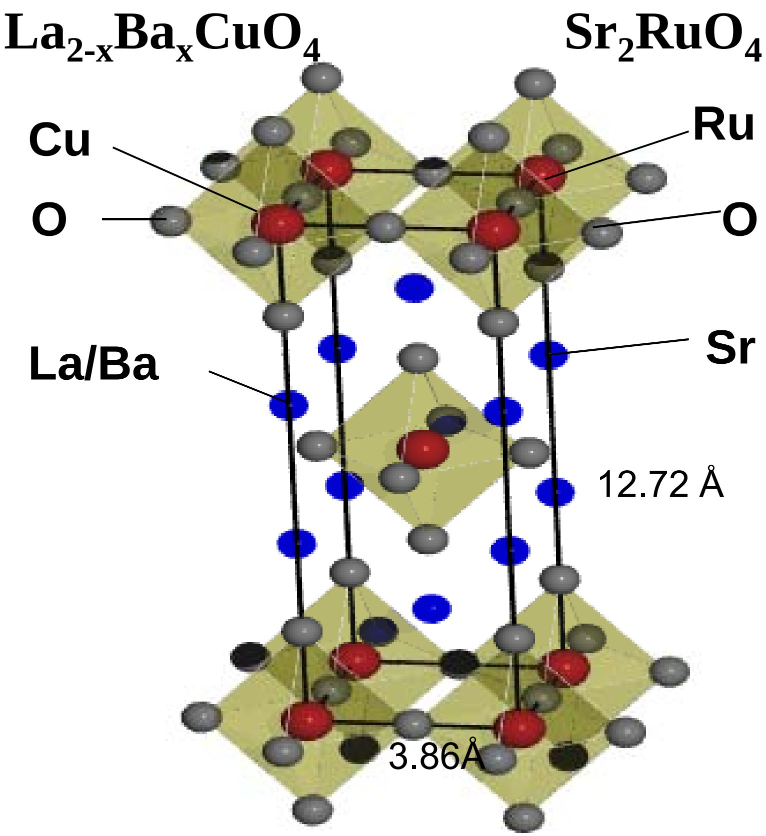
with $$R_0 = V_i V_f \sqrt{\det(A)}\ {}^1\!/\!_{\pi^2}$$ $V_i = p_m k_i^3 \cot(\theta_m)$ et $V_f = p_a k_f^3 \cot(\theta_a)$

V_i provides the same information as the monitor fixed kf

Neutron scattering technique

Application

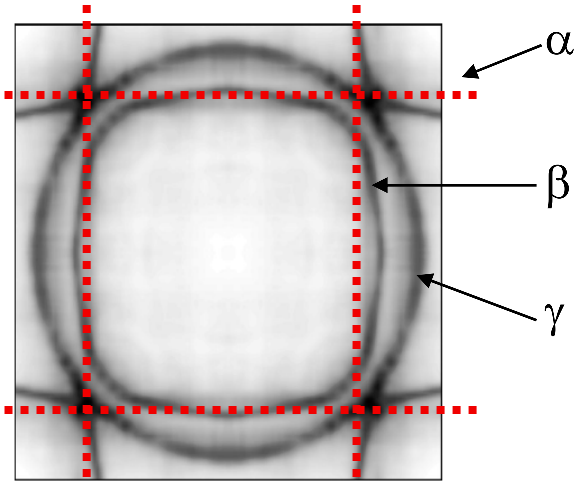
Sr_2RuO_4

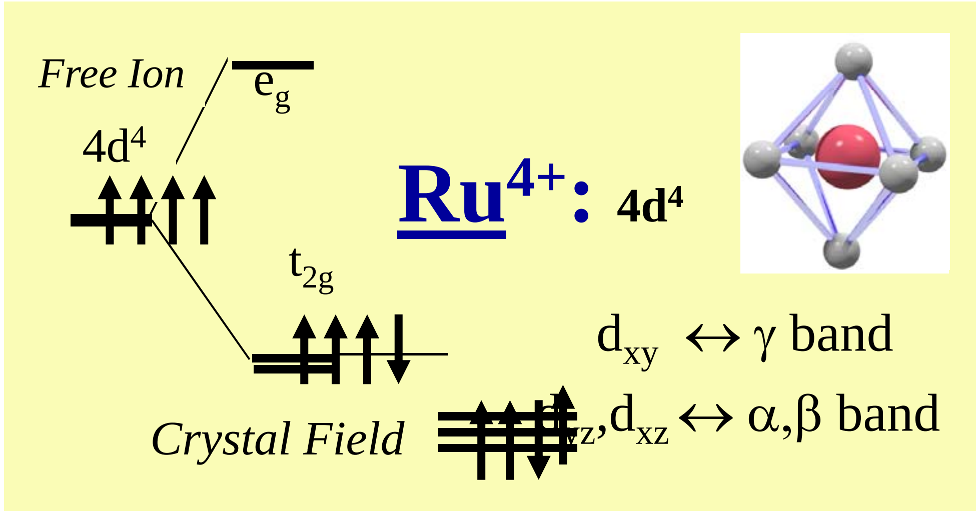


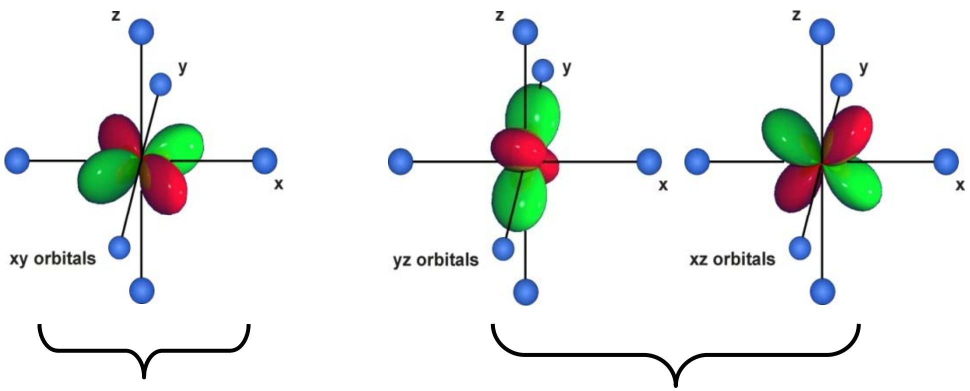
- Superconductivity $T_C \leq 1.5K$ (1994)
- Anisotropic 3-dim. metal $R_C/R_{ab} \approx 450$ $\rho_{ab} < 1 \mu\Omega cm$
- Pauli-Paramagnetism
- Fermi liquid (low T)
- Odd parity (p-wave) Spin-triplet pairing

Sr_2RuO_4 – electronic structure

Fermi surface: 3 sheets

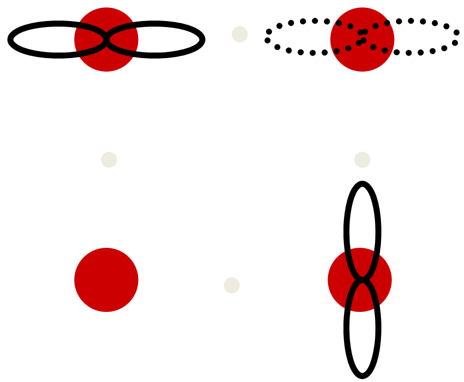






γ (2 dim.)

α, β (1 dim.)

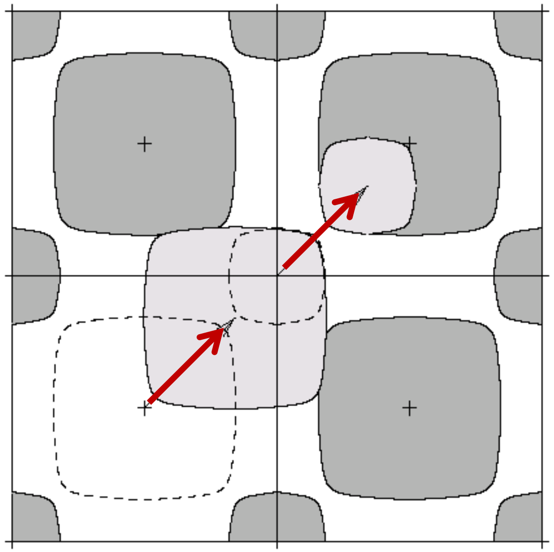


Sr_2RuO_4 – dynamical nesting

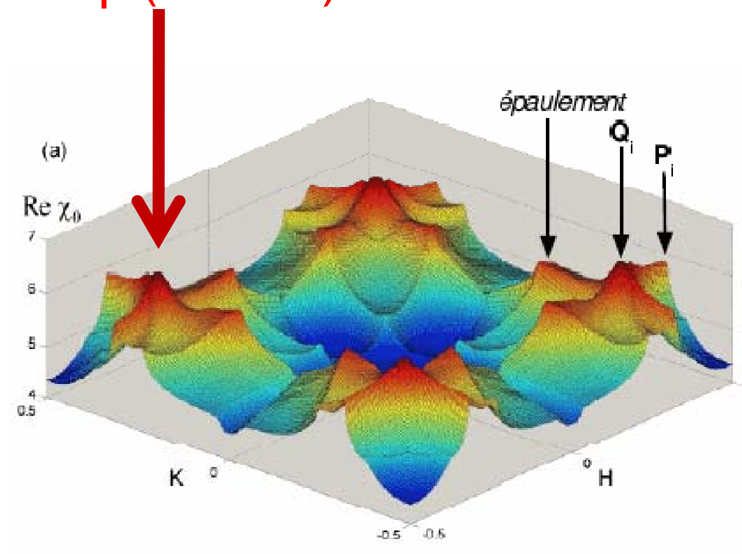
The non interaction spin or charge susceptibility is given by the Lindhardt function

$$\chi_0(q,\omega) = \sum_{k,i,j} \frac{M^{i,j}_{k;(k+q)}[f(\varepsilon_{k,i}) - f(\varepsilon_{(k+q),j})]}{\varepsilon_{(k+q),j} - \varepsilon_{q,i} - \hbar\omega + i0^+} \qquad \begin{matrix} i,j = \alpha,\beta,\gamma \\ M = 0/1 \end{matrix}$$

Strong nesting effect between the quasi-1d α and β bands



Q_i=(0.3 0.3)



unpolarized INS measurements
(single crystals)

Lattice dynamics

Sr_2RuO_4 – lattice dynamics

scattered intensity :

$$I \propto \frac{1}{\omega} \cdot (n(\omega)+1)\{\sum_d \frac{b_d}{\sqrt{m_d}} \cdot e^{(-W_d + i\mathbf{Q}\cdot\mathbf{r_d})} \cdot (\mathbf{Q}\cdot\mathbf{e_d})\}^2$$

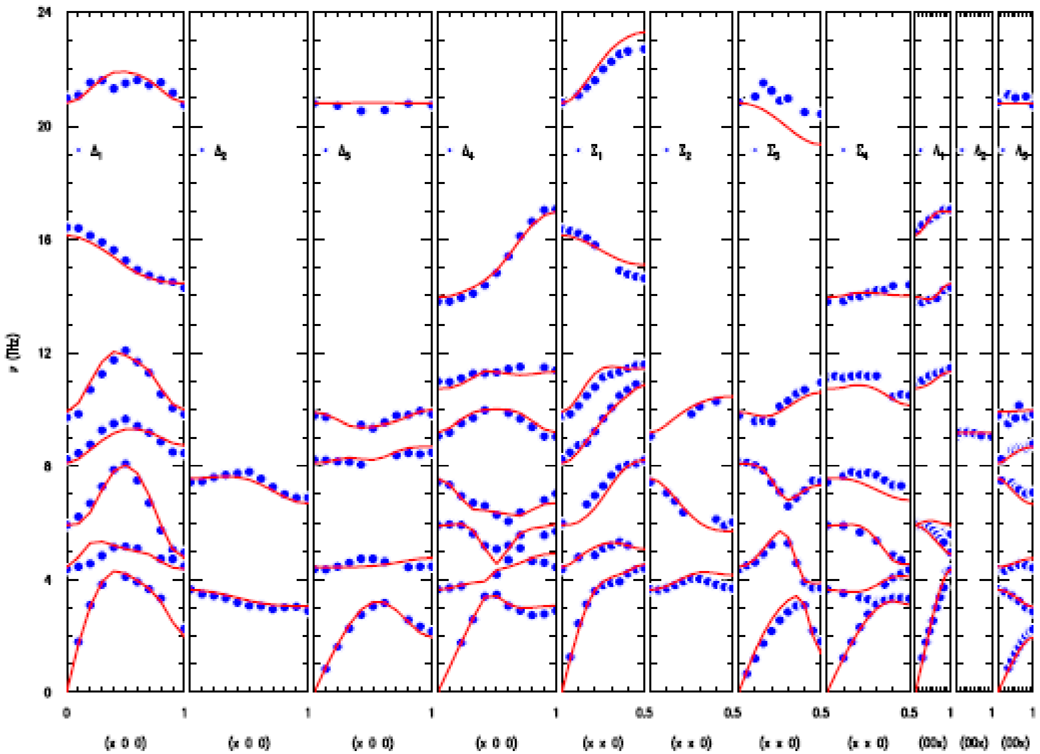
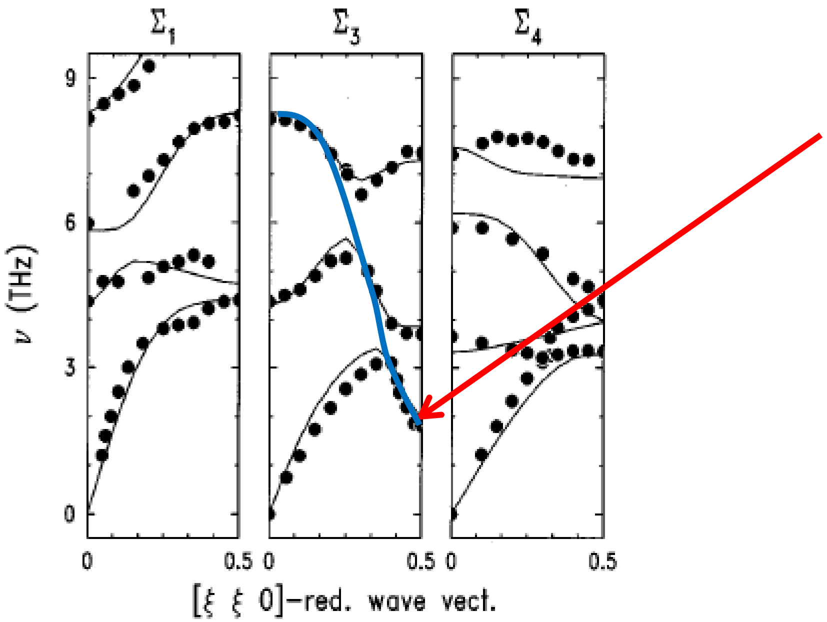


FIG. 2: (color online) Phonon dispersion in Sr_2RuO_4, symbols denote the measured frequencies and lines those calculated with the lattice dynamical model. We show the phonon dispersion along the main symmetry directions, [100], [110] and [001], separated according to the irreducible representations, see text and Table I.

Calculated and measured dispersion curves

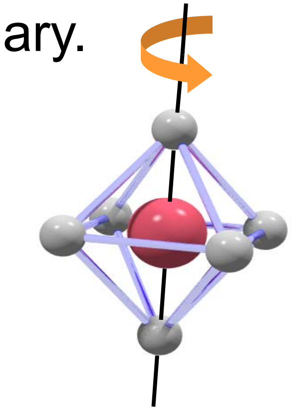
Sr_2RuO_4 – lattice dynamics

Lattice instability:



Σ_3 branch, which ends at the zone boundary (0.5,0.5,0) in the mode corresponding to the RuO_6 octahedron rotation around c axis, exhibits a sharp drop at the zone boundary.

Sr_2RuO_4 is close to rotational instability



Kohn anomaly: No phonon anomaly detected at the planar nesting wave vector Q_i=(0.3,0.3)

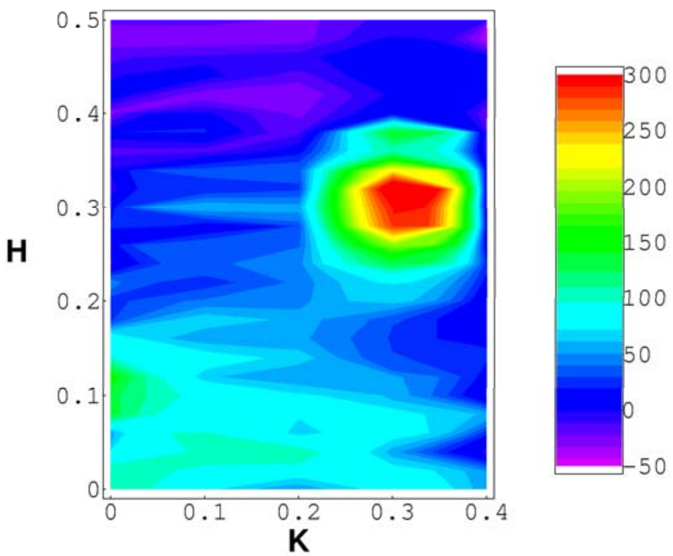
unpolarized INS measurements
(single crystals)

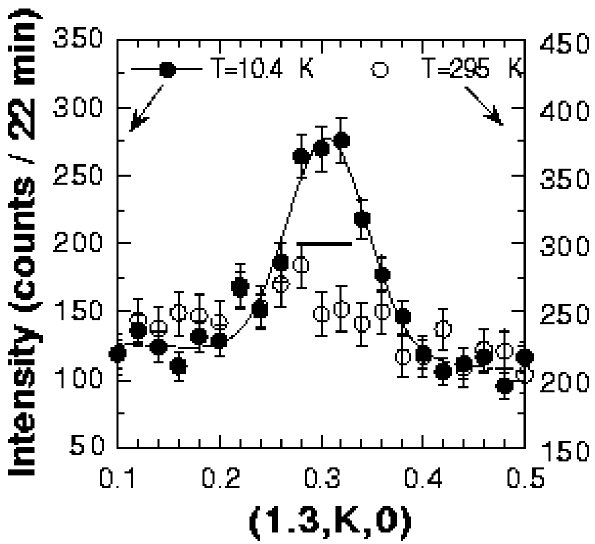
Spin dynamics

Sr_2RuO_4 –spin dynamics

Magnetic scattering:

$$\frac{d^2\sigma}{d\Omega d\omega} = r_0^2 \frac{2F^2(\mathbf{Q})}{\pi(g\mu_B)^2} \frac{\chi''(\mathbf{Q},\omega)}{1-\exp(-\hbar\omega/k_B T)}$$

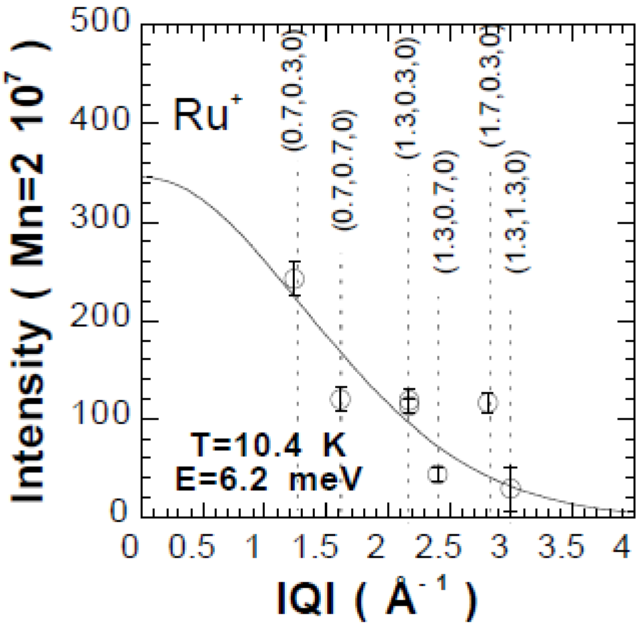


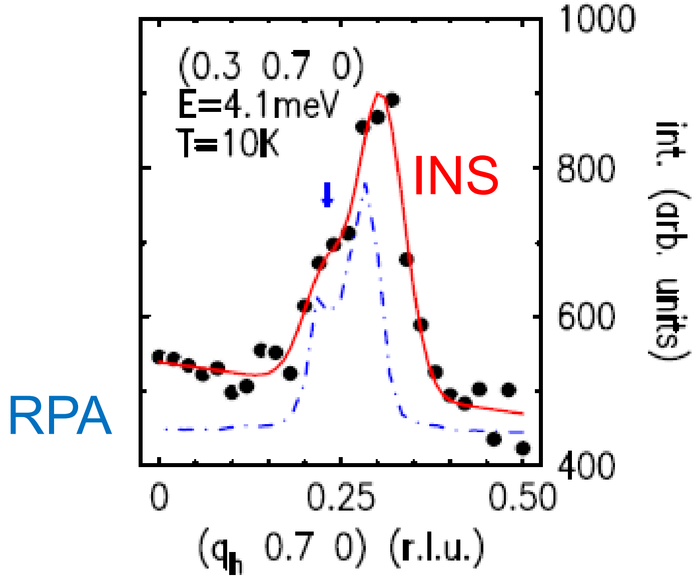


Magnetic response close to the planar nesting wave vector Q_i=(0.3,0.3)

$$\Delta q = 0.13 \pm 0.02 \text{ Å}^{-1}.$$

Sr_2RuO_4 –spin dynamics





2D spin fluctuations

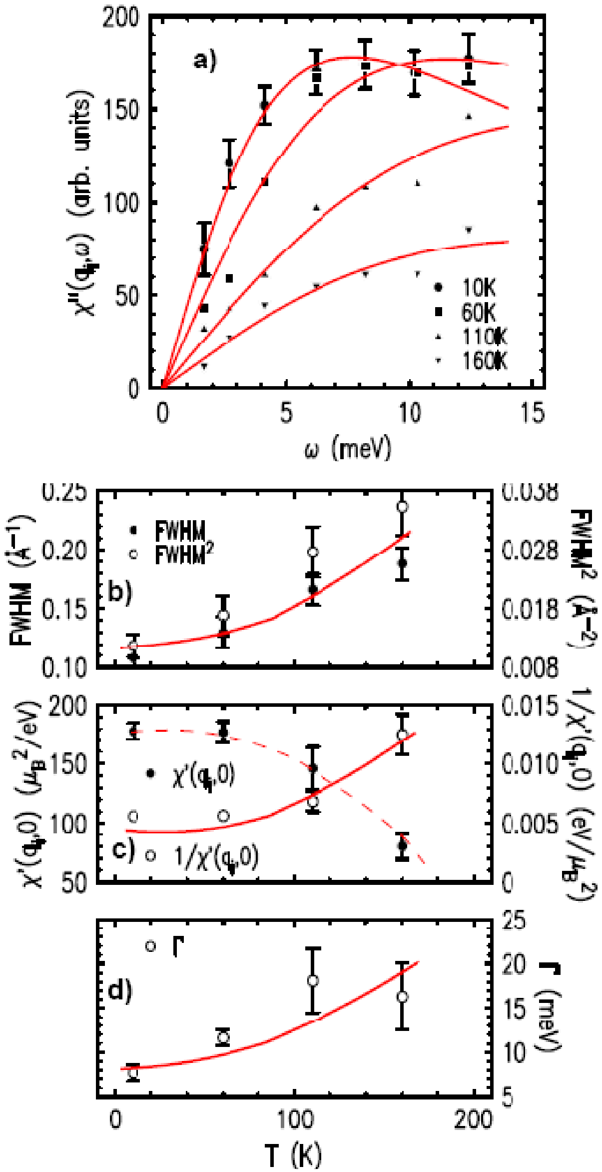
Intensity along c controlled by $F(Q)^2$ only

Intensity decreases at large |Q|

INS measurements in agreement with RPA calculations

$$\chi(q,\omega) = \frac{\chi_0(q,\omega)}{1 - \frac{I(q)}{2(\mu_B)^2}\chi_0(q,\omega)}$$

Sr_2RuO_4 – near a SDW instability



$$\chi''(\mathbf{q_i},\omega) = \chi'(\mathbf{q_i},0)\frac{\Gamma\omega}{\omega^2+\Gamma^2}$$

close to magnetic instability

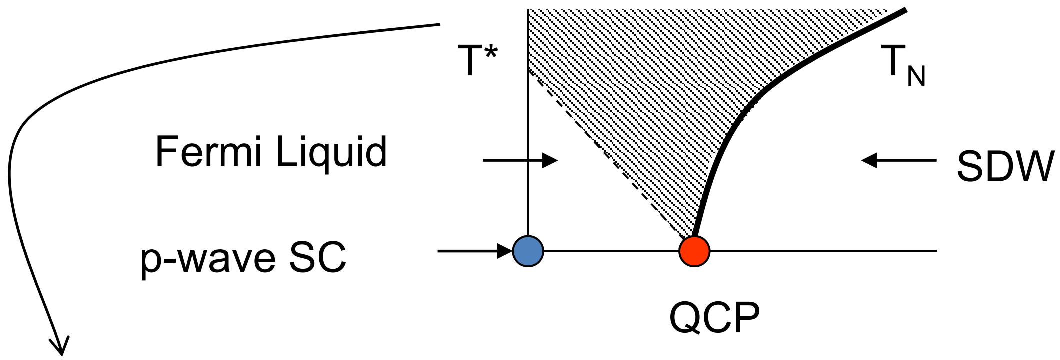
$1-I(q_i)\chi_0(q_i)=\delta \rightarrow 0$

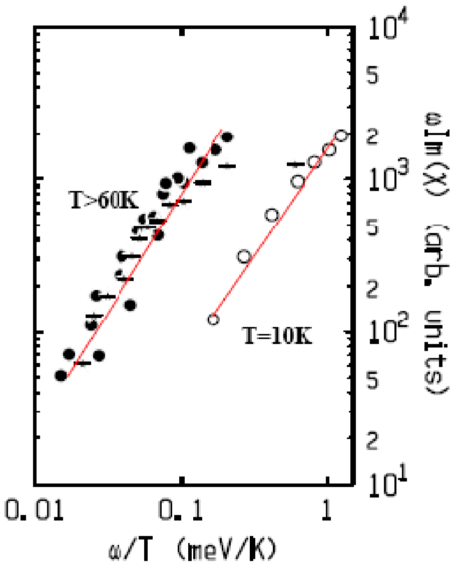
$1/\chi_0(q_i) \sim \Gamma \sim \kappa^2 \sim \delta$

$\delta = 0.03$ at low T

Sidis et al. PRL 83, 3320 (1999)
Braden et al. PRB 66, 064522 (2002)

Sr_2RuO_4 – origin of non Fermi liquid ρ





Braden et al. PRB 66, 064522 (2002)

T>T* ω/T scaling

$$\chi''(\mathbf{q_i}, \omega, T) \propto T^{-\alpha} g(\frac{\omega}{T})$$

Non Fermi liquid : T>T* , ρ ~T

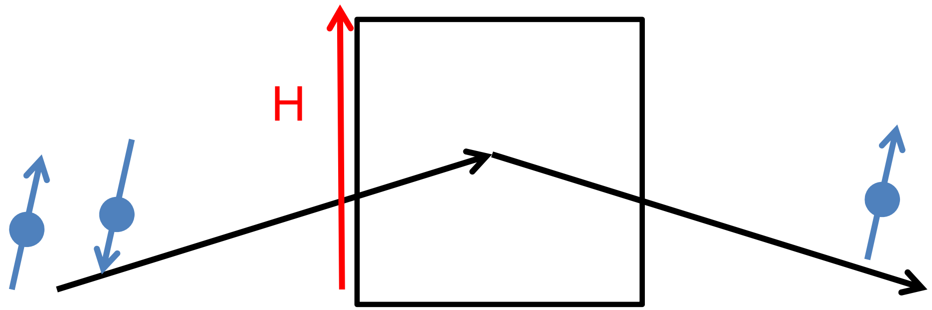
Fermi liquid : T<T* , ρ ~T^2

Polarized INS measurements
(single crystals)

Spin dynamics

Sr_2RuO_4 – polarized INS measurements

Spin polarized neutron beam



Heusler monochromator / analyzer

Single domain ferromagnet

$$|b_N| = |b_m|$$

Interference between nuclear and magnetic scattering

$$| b_N + b_m|^2 = 0$$

$$| b_N - b_m|^2 \neq 0$$

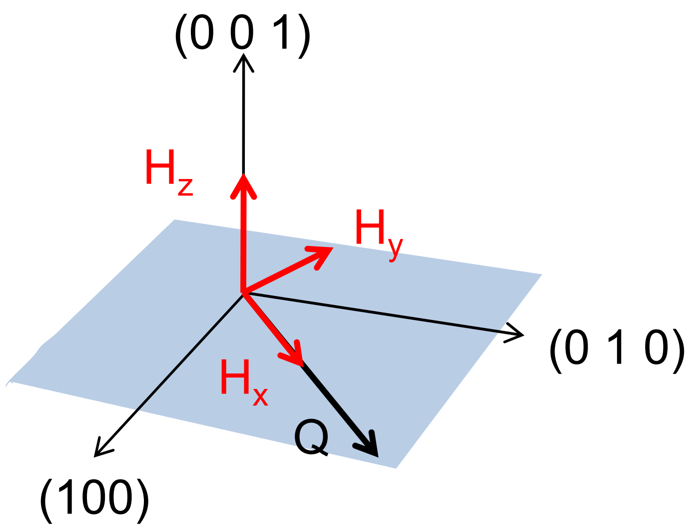
A magnetic guide field keep the spin polarization and defines the polarization P of neutron spin

Sr_2RuO_4 – polarized INS measurements

Polarization:

Nuclear scattering : → non spin flip scattering

Magnetic scattering: spin components: (i) $\perp$Q , (ii) $\perp$P → spin flip scattering



H_x : P // Q

Spin flip channel :

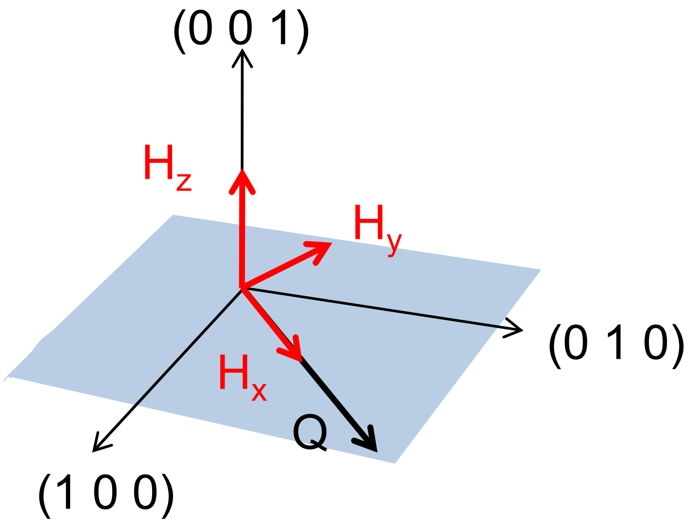
$I_x \sim \text{Im}\,\chi_{a,b} + \text{Im}\,\chi_c + \text{Bg}$

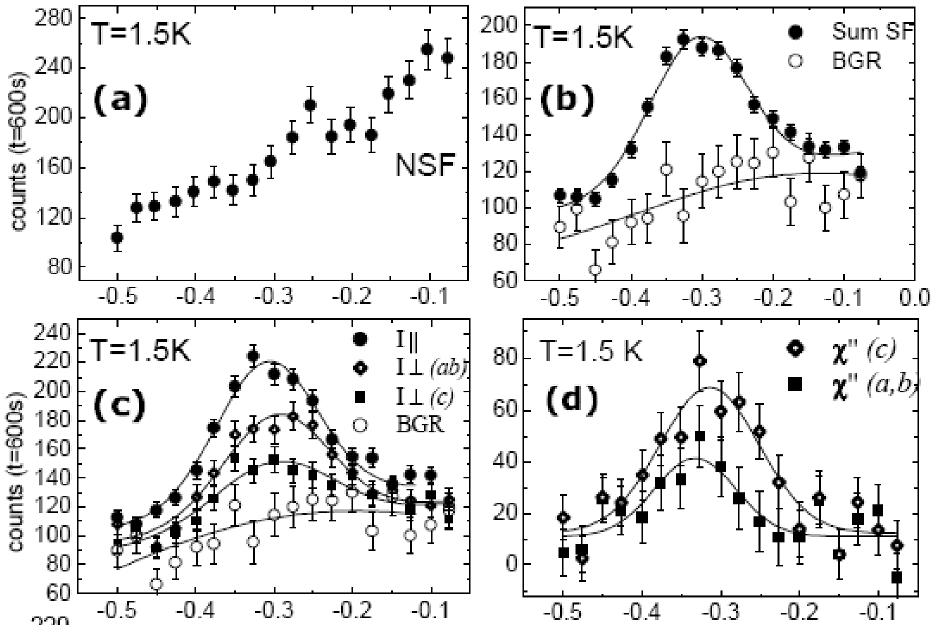
$I_y \sim \text{Im}\,\chi_c + \text{Bg}$

$I_z \sim \text{Im}\,\chi_{a,b} + \text{Bg}$

Sr_2RuO_4 – spin anisotropy

Polarized neutron   8 meV





H_x : P // Q

Anisotropy of IC spin fluctuations at Qi

$\chi_c(q_i) / \chi_{a,b}(q_i) \sim 1.6$

spin-orbit coupling

\+ strong correlation

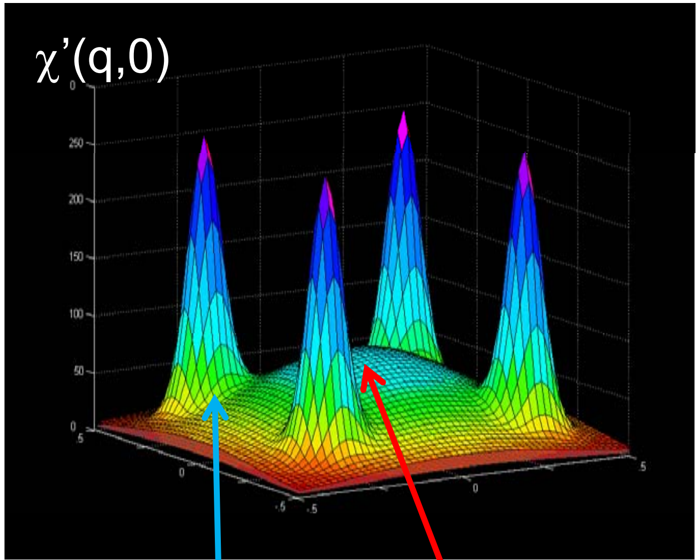
$\chi_c(q_i) / \chi_{a,b}(q_i) > 1$

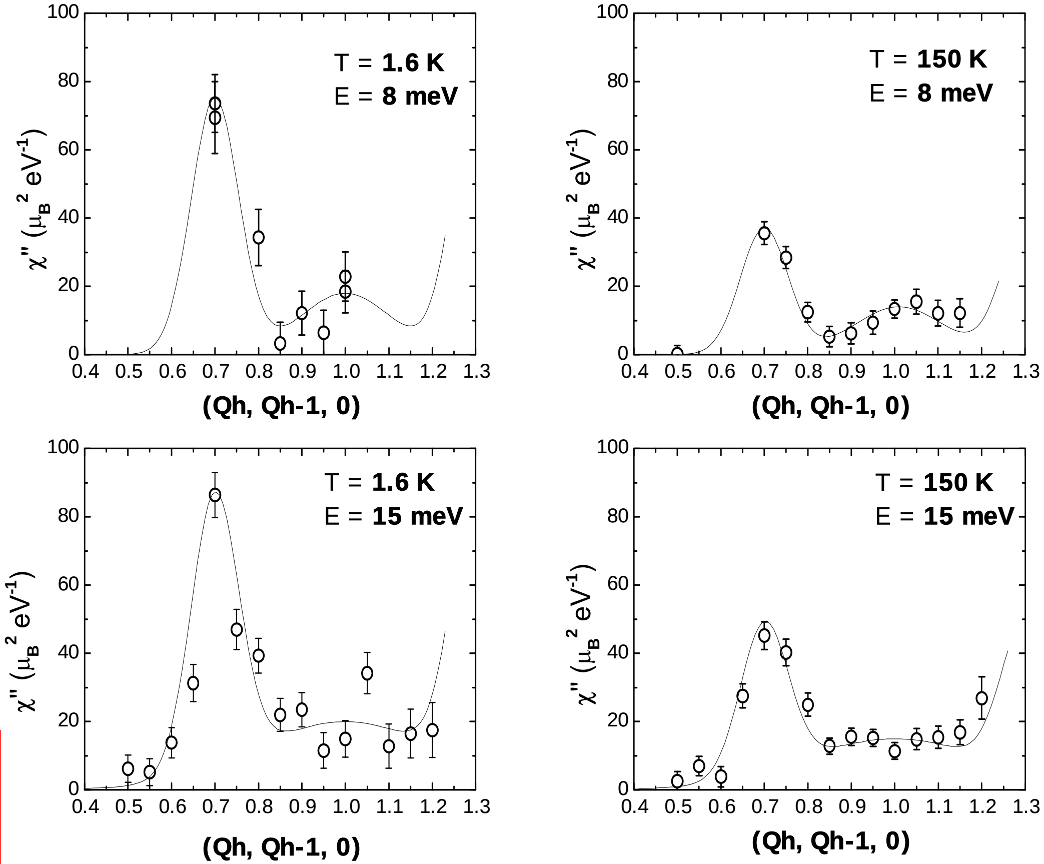
anisotropic spin fluctuations can favor spin tripet SC

Ng et al. J. Phys. Soc. Jpn, (2002).
Manke et al., PRB (2002).

Sato et al. J. Phys. Soc. Jpn, (2000).
Kawabara et al., PRB (2000).

Sr_2RuO_4 – full spin susceptibility





Broad and weak excitations around Q=0 (Γ~20 meV)

γ band

sharp and strong excitations around Qi (Γ~7 meV)

α, β bands

H_x : P // Q – spin flip channel

INS measurements

Versus

NMR
Specific heat
ARPES

Sr_2RuO_4 – NMR – local probe

INS

Spin-lattice relaxation

$$(1/T_1T) \simeq \frac{k_B\gamma_n^2}{(g\mu_B)^2}\sum_q |A(\mathbf{q})|^2 \left.\frac{Im\chi(\mathbf{q},\omega)}{\omega}\right|_{\omega\to 0}$$

Hyperfine coupling

$$^{17}\mathrm{O},\ |A(\mathbf{q})|^2 = \Lambda^2[1+1/2(\cos(\tfrac{2\pi}{a}q_x)+\cos(\tfrac{2\pi}{a}q_y))]\ (\Lambda = 33\ \mathrm{kOe}/\mu_B\quad)$$

$$^{101}\mathrm{Ru},\ A(\mathbf{q}) = -299\ \mathrm{kOe}/\mu_B$$

1.6 K		$^{101}(1/T_1T)$	$^{17}(1/T_1T)$	150 K		$^{101}(1/T_1T)$	$^{17}(1/T_1T)$
	FM	5.6	0.33		FM	4.9	0.29
	IC	12.2	0.38		IC	7.6	0.25
	FM+IC	17.8	0.71		FM+IC	12.4	0.54
	NMR	15	0.8		**NMR**	8.5	0.45

Sr_2RuO_4 – specific heat

$$\gamma = C/T$$

Spin fluctuation contribution
At low temperature

$$\gamma_{sf} = \frac{1}{N}\frac{C_{sf}}{T} = \frac{1}{N}\frac{\pi k_B^2}{\hbar}\sum_{\mathbf{q}}\frac{1}{\Gamma(\mathbf{q})}$$

$$37.5\ \frac{J.meV}{mol.K^2}$$

IC: $4\frac{J.meV}{mol.K^2}$

FM: $33\frac{J.meV}{mol.K^2}$

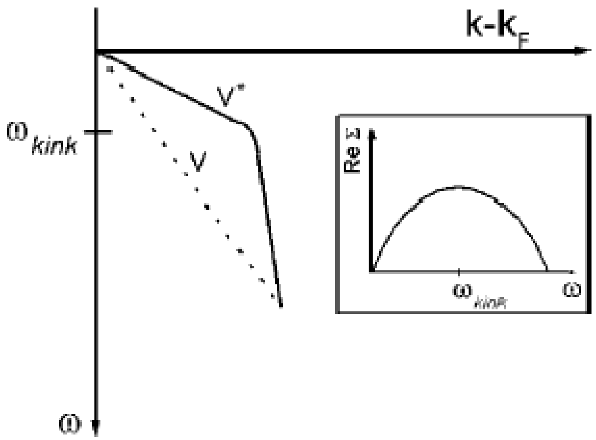
Sr_2RuO_4 – charge excitations - ARPES

ARPES : spectral function of quasiparticles $A(k,\omega) = -\frac{1}{\pi} Im G(k,\omega)$

Scattering : self-energy $G(k,\omega)^{-1} = G_o(k,\omega)^{-1} - \Sigma(k,\omega)$

$$\Sigma(k,\omega) = \frac{1}{\pi^2}\frac{1}{N}\sum_q \int_{-\infty}^{\infty} d\Omega d\nu V_{qk}^2 Im\chi(q,\Omega) Im G(k+q,\nu) \times \{\frac{n(\Omega)+f(\nu)}{\omega+\Omega-\nu+i\epsilon}\}$$

collision with bosonic modes (phonons, spin excitations,....)



This leads to the "kink condition"

$$\omega_{kink} \approx E_{\mathbf{k-Q}} + \Omega \quad .$$

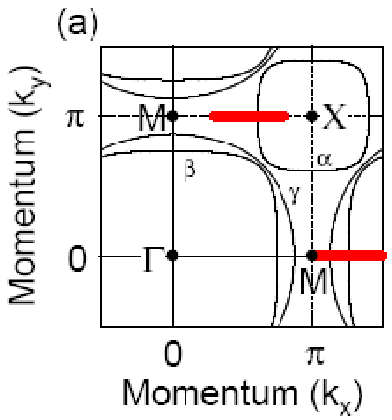
Ω : characteristic energy of bosonic modes (phonons or spin excitations)

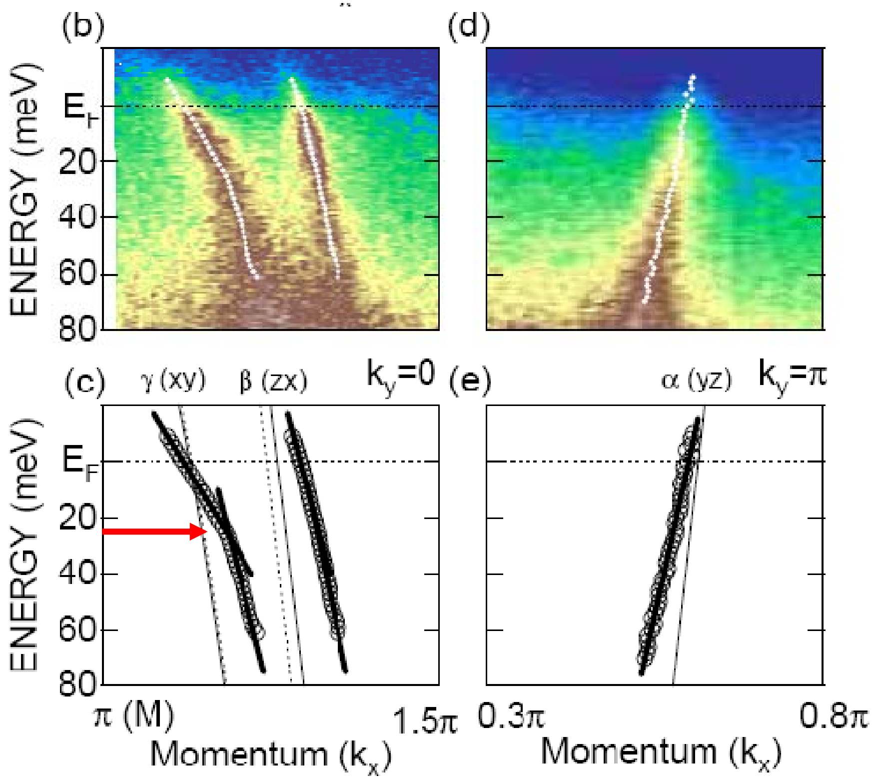
Manske et al., PRB 67, 134520 (2003)
Manske et al., PRB 65, 220502 (2002)

Sr_2RuO_4 – charge excitations - ARPES

Iwasawa et al., condmat/0508312

ARPES





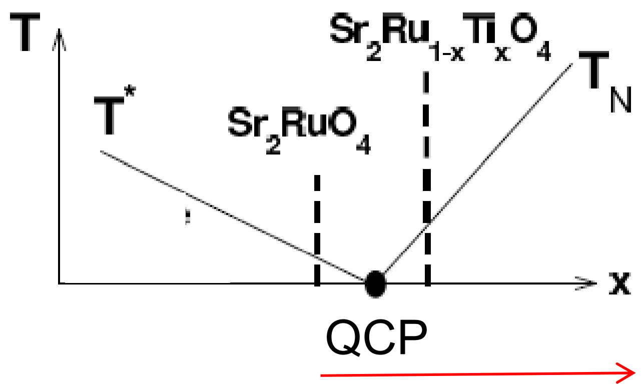
observed *Kink* in the γ band
ω_{sf}~25meV

Role of the *pseudo-FM* fluctuations ?...... Role of phonons?

Unpolarized elastic measurements
(single crystal)

Magnetic order

Sr_2RuO_4 – order from disorder



Non magnetic Ti^{4+}

Magnetic wave vector q_0

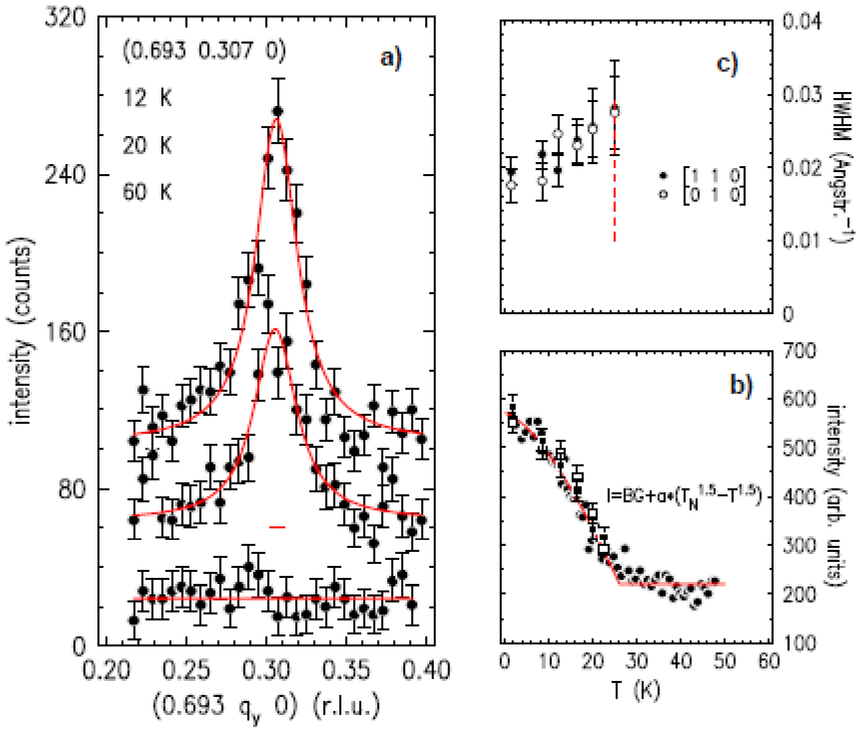
$$\left(\frac{d\sigma}{d\Omega dE'}\right)_{elas} \propto F(Q)^2[\frac{1}{2}(1+\frac{L^2}{Q^2})m_{a,b}^2+(1-\frac{L^2}{Q^2})m_c^2]\frac{1}{(\vec{q}-\vec{q}_O)^2+\Delta q^2}\delta(\vec{Q}-\vec{q}-\vec{\tau})\delta(\omega)$$

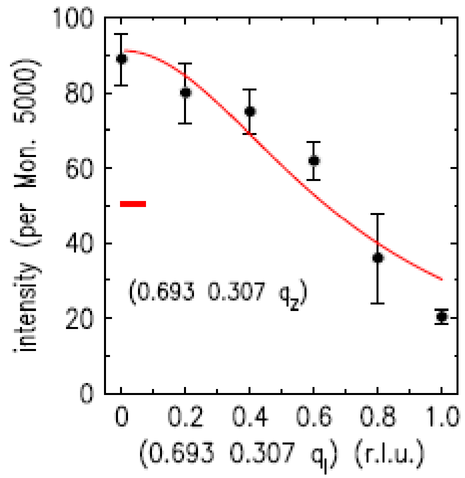
Intensity proportional to the square of the order moment

Orientation factor gives the direction of the order moment

Correlation length $\Delta q \sim \xi^{-1}$

Sr_2RuO_4 – order from disorder





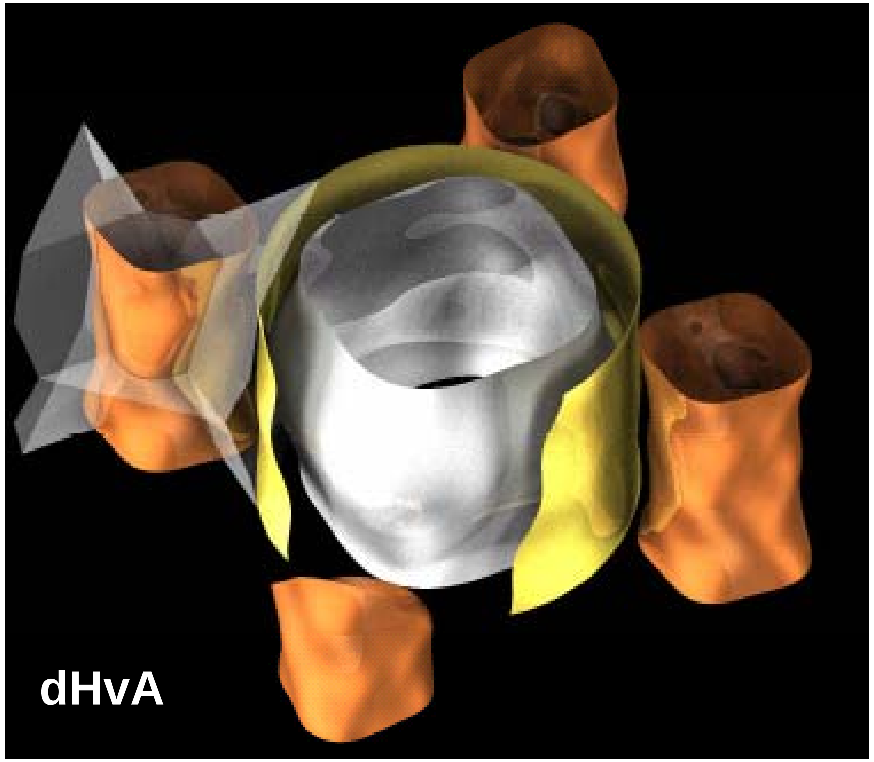
Study : Q= q_o+ τ , q_o =(0.307,0.307,1) , τ = (1 0 -1) [I4mmm structure]

Ordered moment: m = 0.3 μ_B Néel temperature : T_N = 25 K

Planar correlation length : $\xi_{a,b}$ ~50 Å

along c : hardly correlated system

Sr_2RuO_4 – mag. Order vs Fermi surface



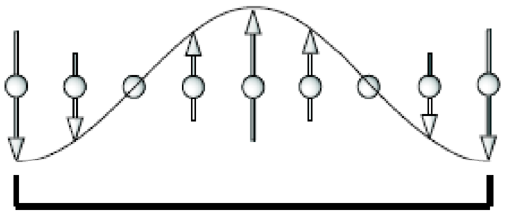
C. Bergemann et al.

q_o =(0.307,0.307, 1)

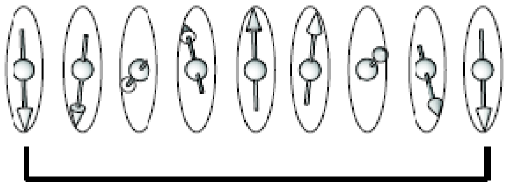
The order wave vector is 3D

Modulation along c of the α and β (involved in nesting)

Sr_2RuO_4 – spin density wave



M1 : SDW with m // c



M4 : helimagnet

(hkl)-indices	M1	M2	M3	M4	observed
(0.307 0.307 1)	1	1	1	1	1
(0.307 0.693 0)	0.61	0.05	1.10	0.28	0.51(4)
(0.307 0.307 3)	0.05	0.19	0.46	0.13	0.08(3)
(0.693 0.693 1)	0.23	0.17	0.05	0.20	0.27(5)
(0.693 0.693 3)	0.06	0.07	0.10	0.07	0.0(1)
(1.307 0.307 0)	0.11	0.05	0.06	0.07	0.17(5)
(1.307 0.693 1)	0.07	0.01	0.14	0.06	0.07(2)
(0.307 0.307 5)	0.00	0.04	0.12	0.03	0.00(4)

FULLPROF- program

magnetic Moments are // to c
consistent with the spin anisotropy observed in Sr_2RuO_4

Origin: spin-orbit coupling

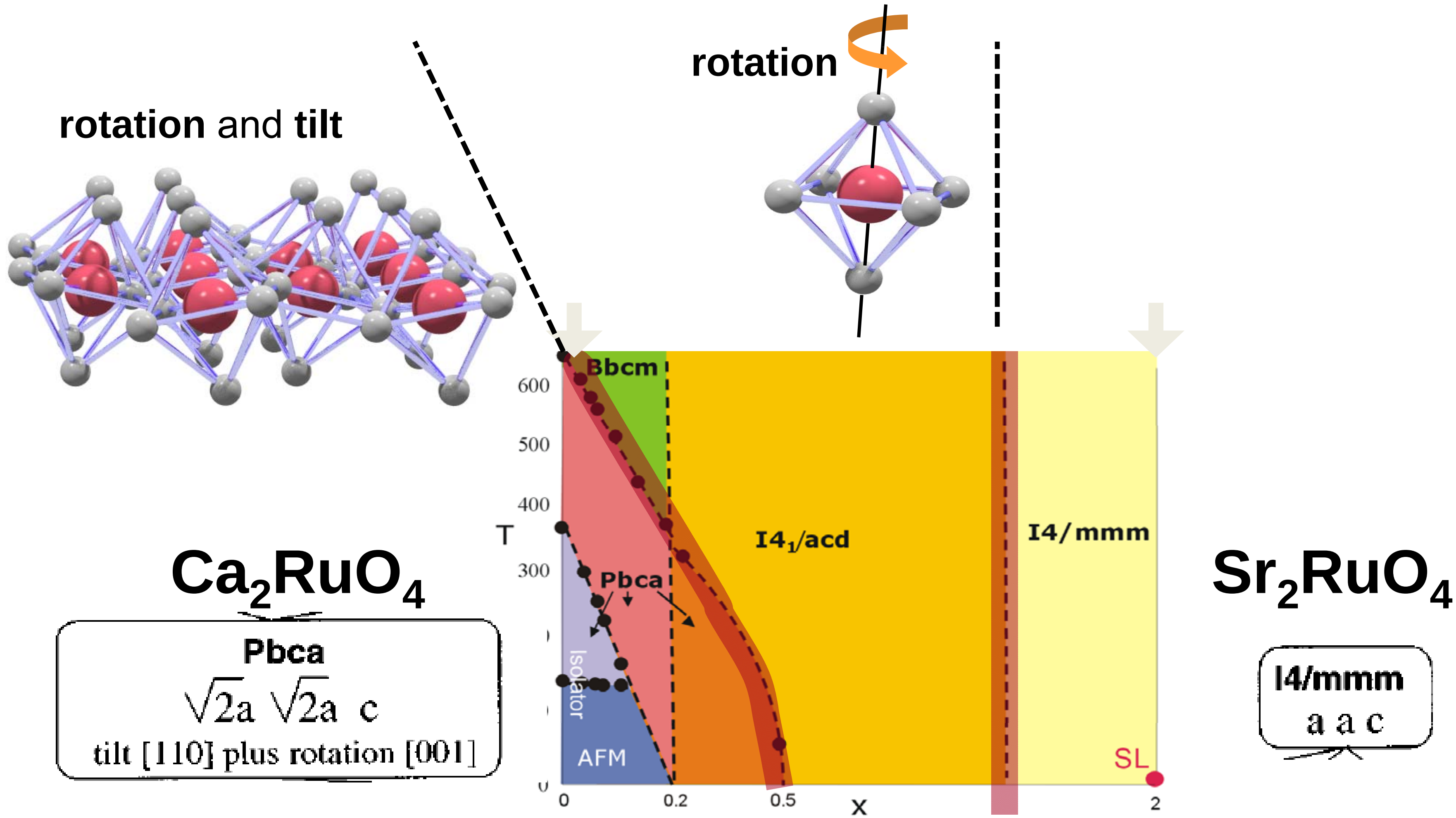
Powder diffraction measurements

Magnetic and nuclear structures

Phase diagram

$Ca_{2-x}Sr_xRuO_4$: Structural properties

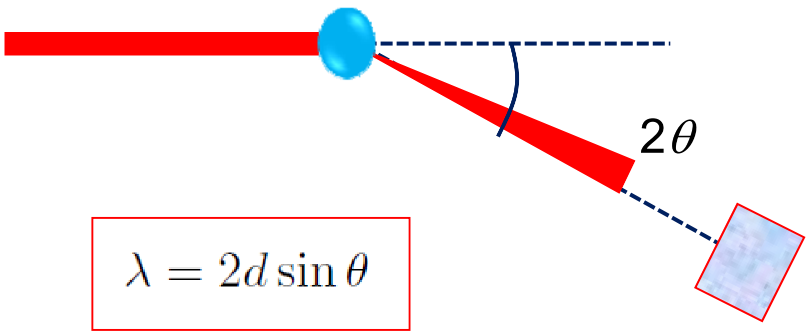
Isovalent substitution $r_{Ca} < r_{Sr}$ Ca^{2+} r = 1.18Å ↔ Sr^{2+} r = 1.31Å



Nakatsuji et al. PRL 84 2666 (2000), Friedt et al. PRB 63, 174432 (2001), Braden et al. PRB 58, 847 (1998)

Ca_2RuO_4: Nuclear structure

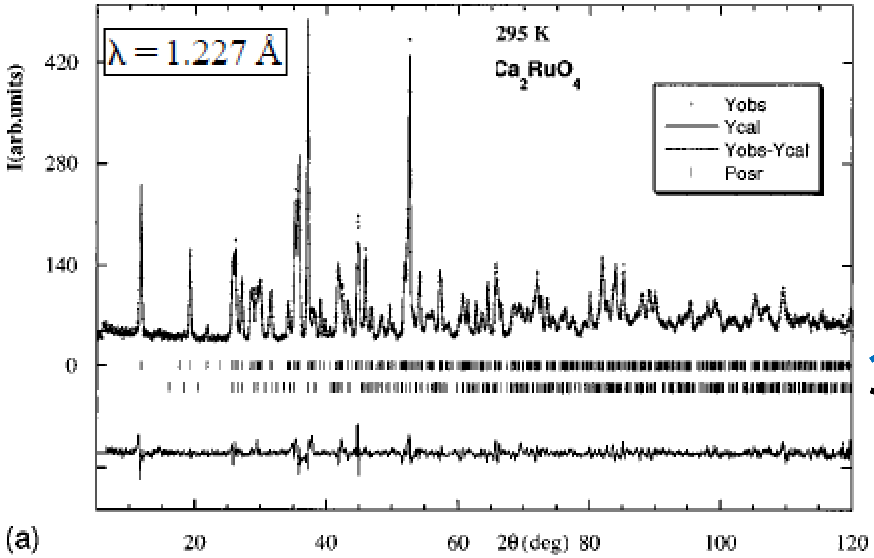
Incident neutron beam



$$\lambda = 2d\sin\theta$$

$$\begin{aligned} Q^2 &= k_i^2 + k_f^2 - 2k_i k_f \cos 2\theta \\ &= 2k_i^2\,(1-\cos 2\theta) \\ &= 4k_i^2 \sin^2\theta \\ Q &= 2k_i \sin\theta \end{aligned}$$

$$\frac{2\pi}{d} = 2\frac{2\pi}{\lambda}\sin\theta$$



Bragg reflections : Ca_2RuO_4

Bragg reflections: $CaRuO_3$ (*impurities*)

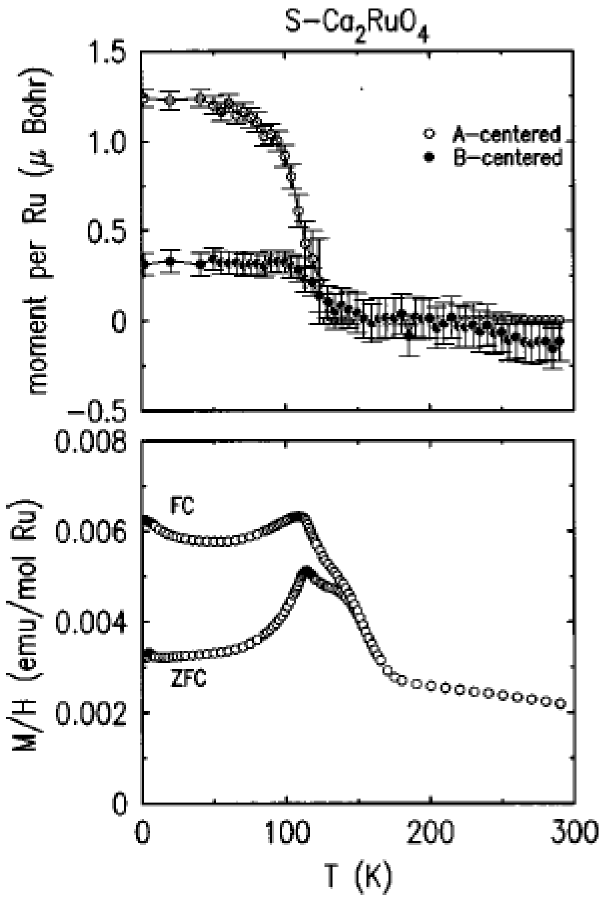
Friedt et al. PRB 63, 74432 (2001),
Braden et al. PRB 58, 847 (1998)

Ca_2RuO_4: Magnetic structure

Mott Insulator Ca_2RuO_4 : **T<Tn, A-type AF order**

Magnetic moments // b = elongation of octahedra

Magnetic moment ~1.3 μ_B < 2μ_B (localized S=1 , Ru^{4+})



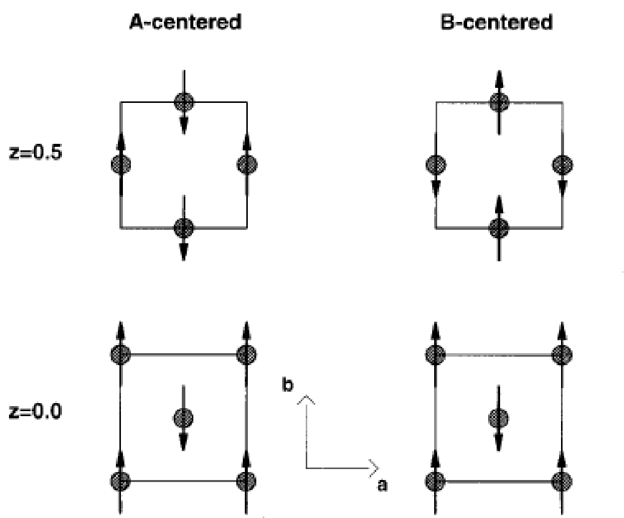


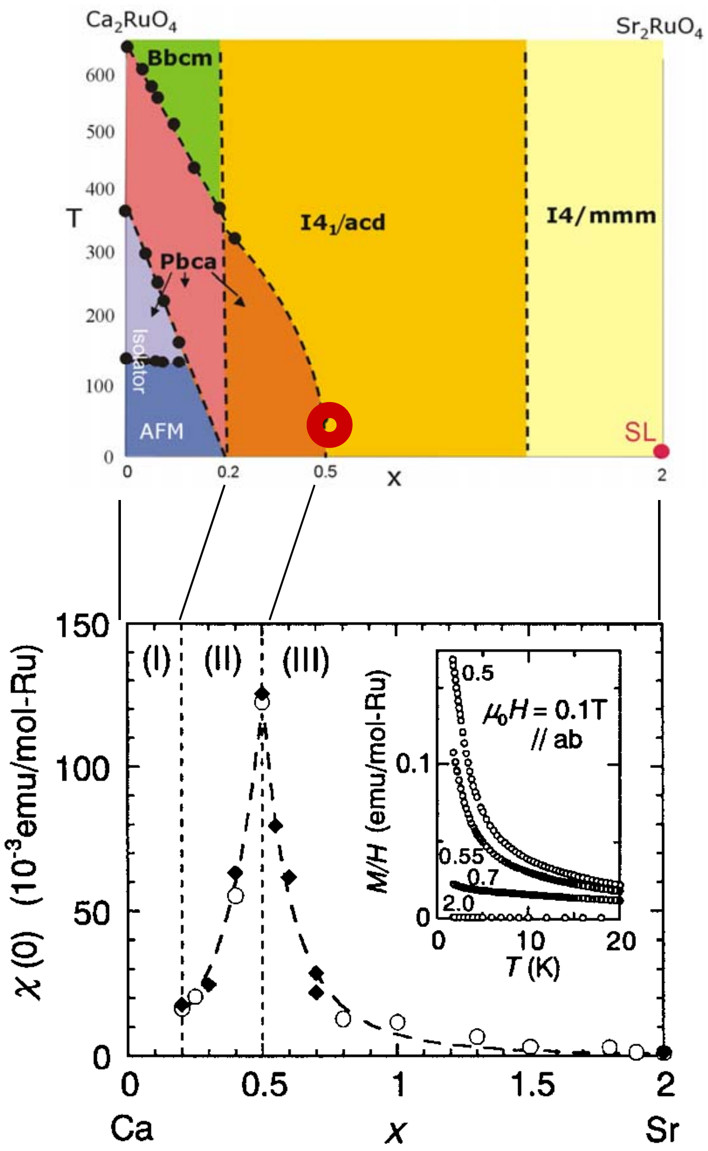
FIG. 10. Schematic picture of the two magnetic modes in space group *Pbca*. Only the spin directions of the Ru's at $z=0.0$ and $z=0.5$ are shown. The propagation vector of the *A*-centered mode is (1 0 0) (La_2CuO_4 type) and that of the *B*-centered type is (0 1 0) (La_2NiO_4 type).

Friedt et al. PRB 63, 74432 (2001),
Braden et al. PRB 58, 847 (1998)

Polarized neutron diffraction
(high quality sample)

Spin density maps

$Ca_{2-x}Sr_xRuO_4$: meta-magnetism



Nakatsuji et al. PRL 90 (2003)

Intermediate Sr Content

$0.2 < x < 0.5$

<u>Metallic state</u>

- near magnetic instability
- high specific heat
- metamagnetic transition

- octahedra tilt

↓

<u>Band structure :</u>

narrowing of the γ band

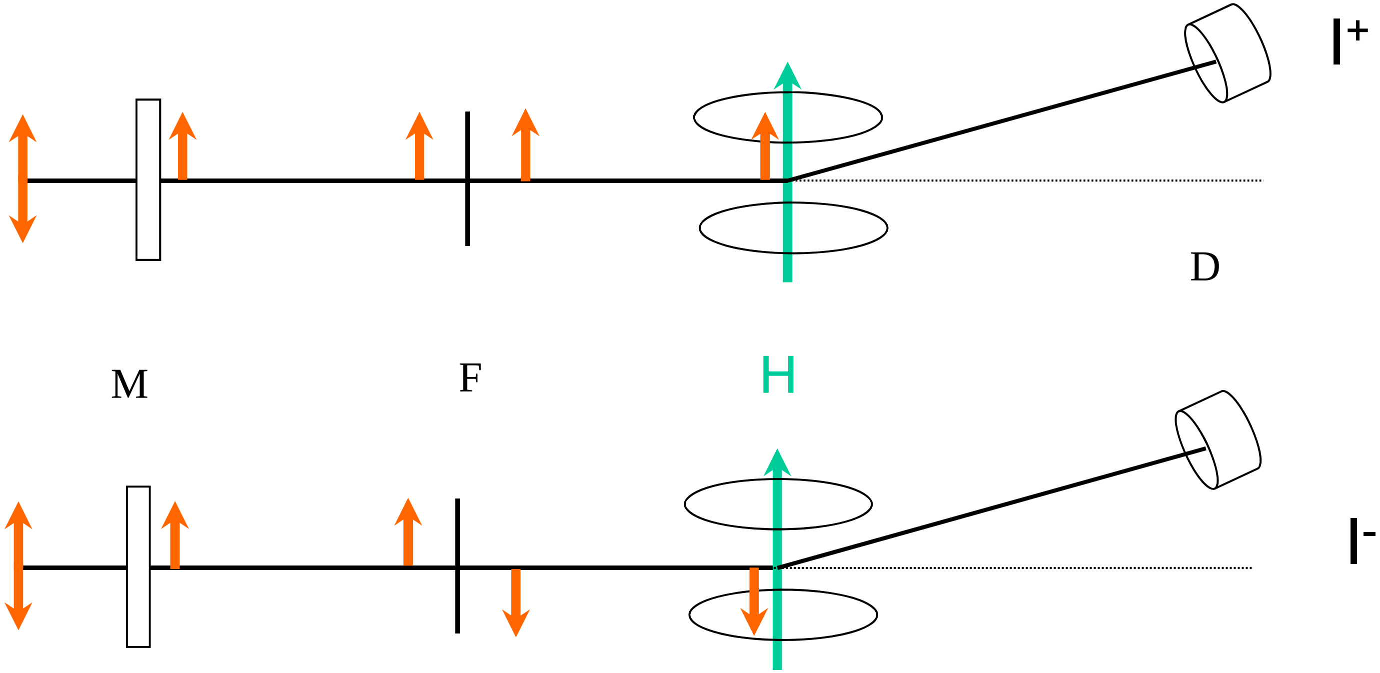
van Hove singlarity shifts below Ef

↓

<u>Hall measurements</u>

Sign change of charge carriers

$Ca_{2-x}Sr_xRuO_4$: spin density map



$$R = \frac{I_+}{I_-} = \left(\frac{F_N + F_M}{F_N - F_M} \right)^2$$

$$I^{\pm} \propto (F_N \pm F_M)^2$$

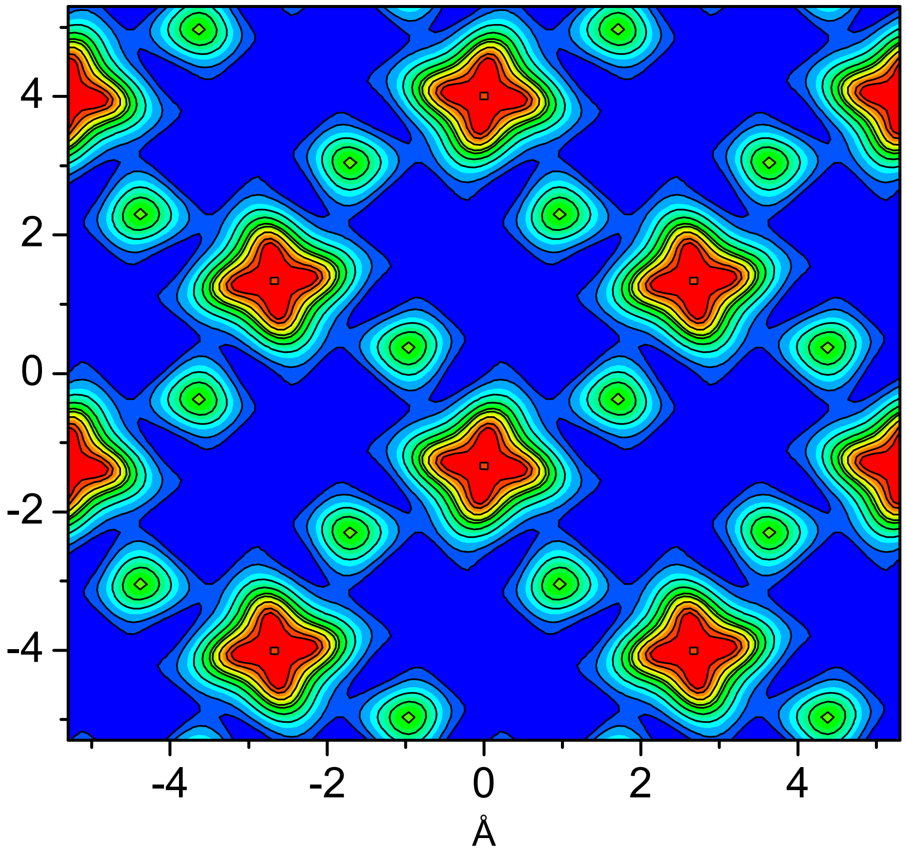
$$F_M << F_N$$

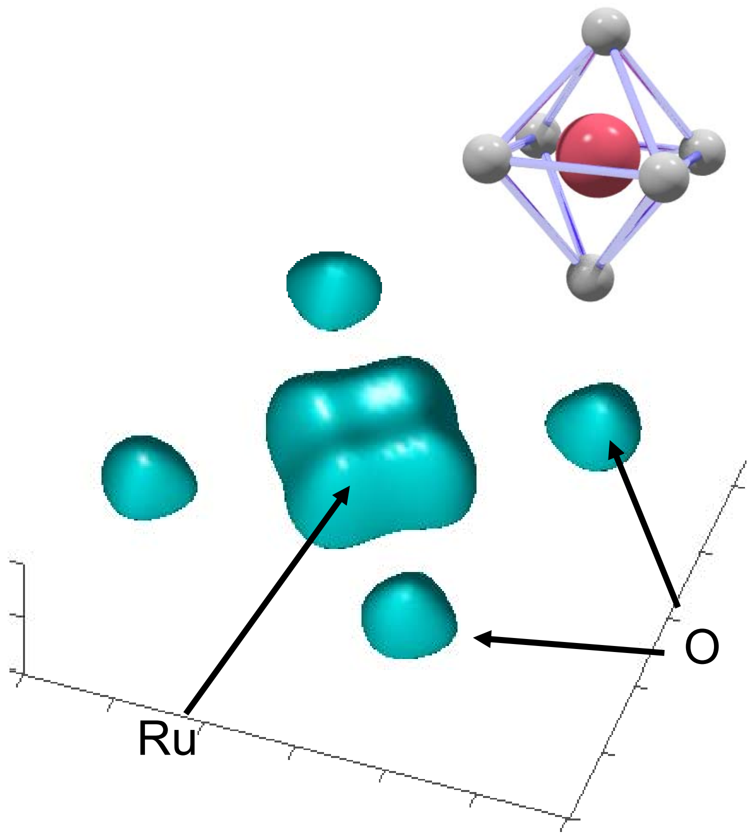
$$R \cong 1 + 4 \frac{F_M}{F_N}$$

$Ca_{2-x}Sr_xRuO_4$: spin density map at x=0.5

0.35 μ_B Ruthenium ⟶ $4d_{xy}$ orbitals (γ band)
0.08 μ_B Oxygen (in-plane) ⟶ significant amount of magnetization on oxygen
0.01 μ_B Oxygen (apical)

RuO_2 plane:





Inelastic neutron scattering
(single crystal)

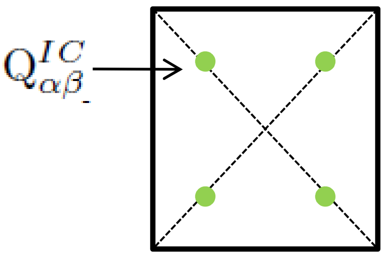
External magnetic field

$Ca_{1.8}Sr_{0.2}RuO_4$: meta-magnetism

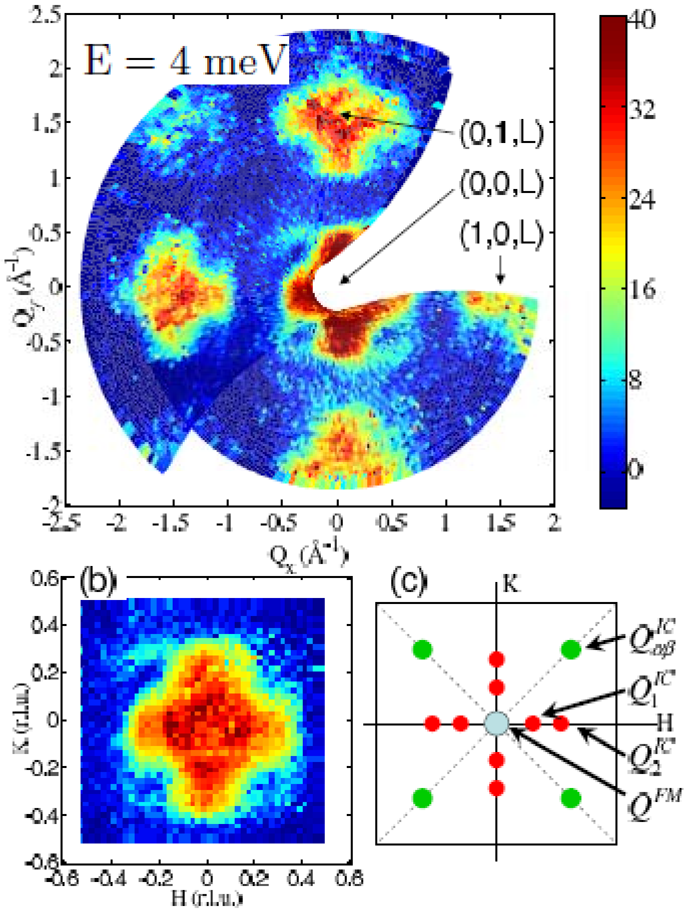
INS:

$$\frac{d^2\sigma}{d\Omega d\omega} \propto \frac{F^2(\mathbf{Q})}{1-\exp(-\frac{\hbar\omega}{k_B T})} \cdot \chi''(\mathbf{Q},\omega)$$

Sr_2RuO_4

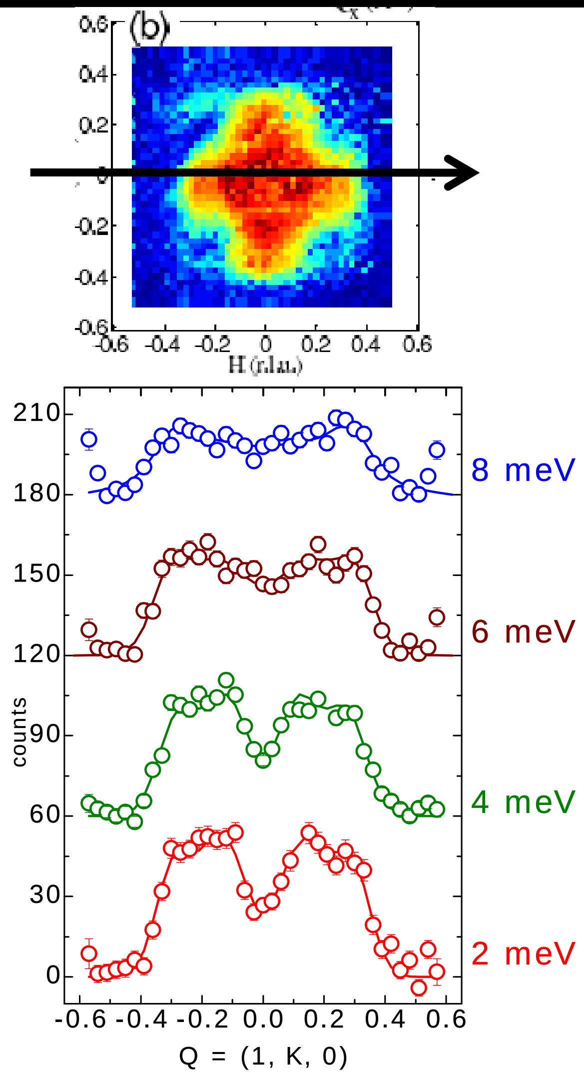


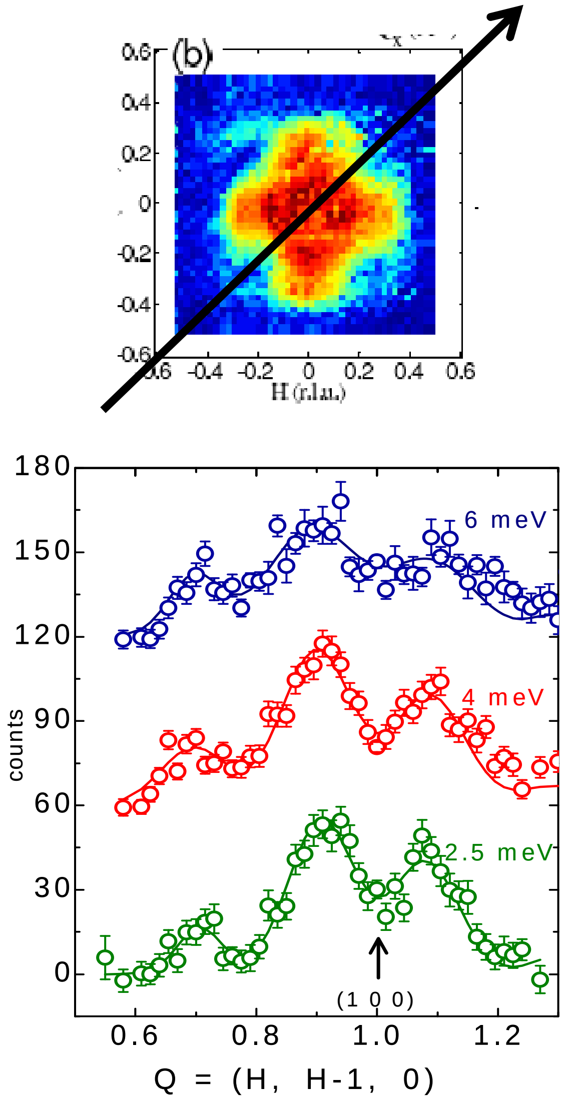
$Ca_{1.8}Sr_{0.2}RuO_4$



Steffens et al. Cond-mad(2010)

$Ca_{1.8}Sr_{0.2}RuO_4$: meta-magnetism

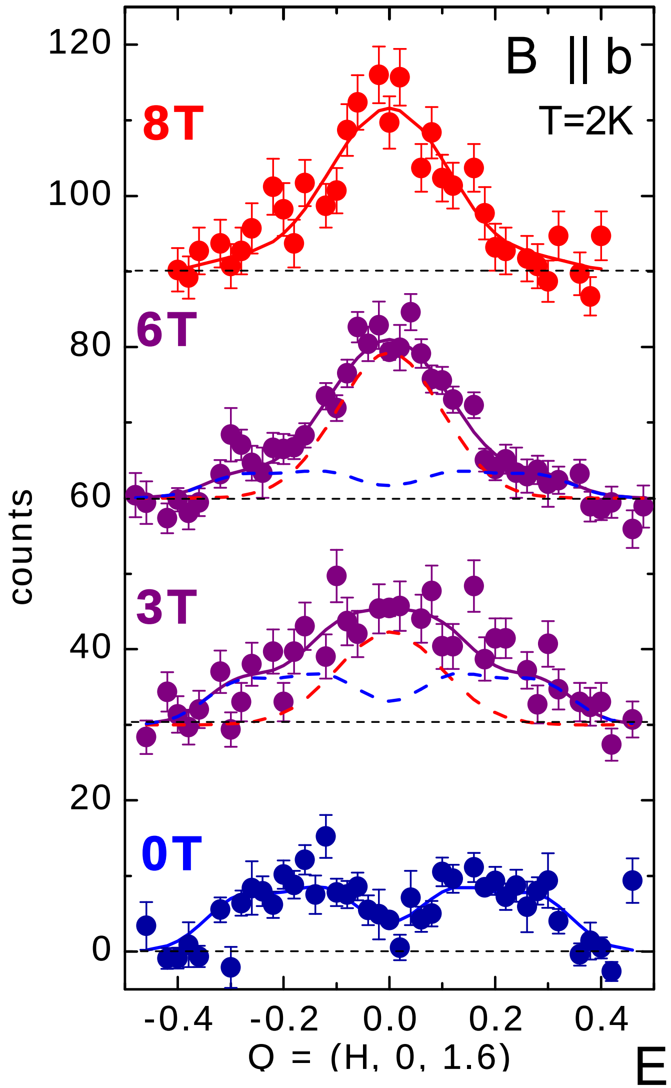




contributions from several **Q**
(q_1=0.12, q_2=0.27)

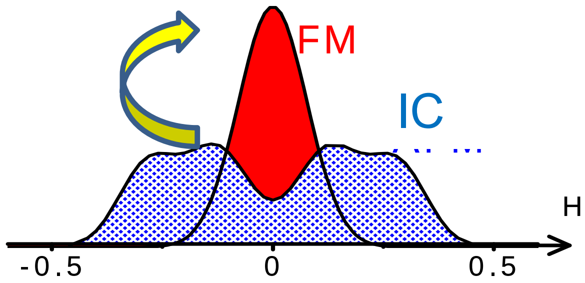
Steffens et al. Cond-mad(2010)

$Ca_{1.8}Sr_{0.2}RuO_4$: meta-magnetism



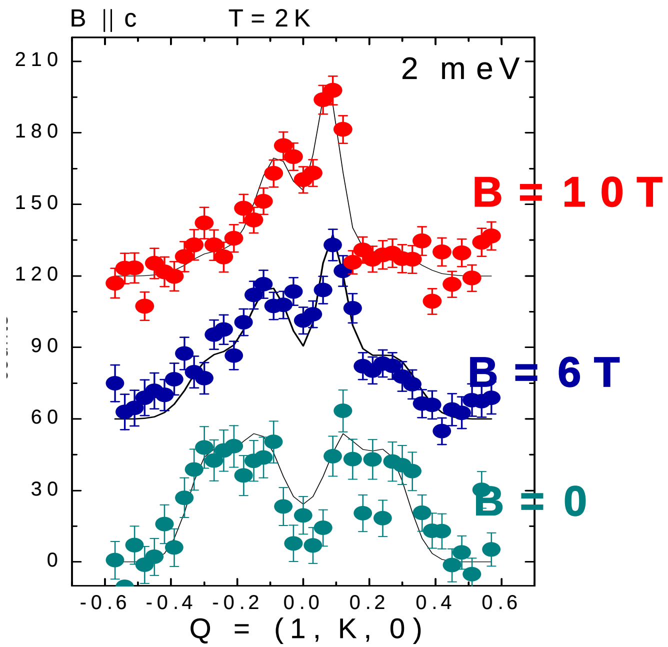
E=2meV

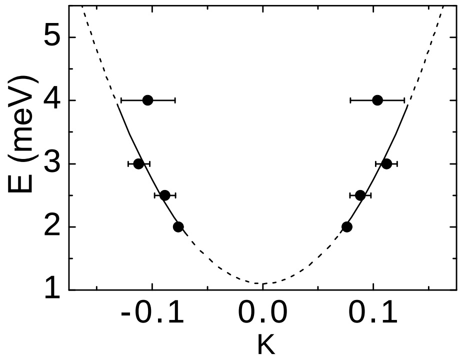
Fundamental change of magnetic fluctuations at metamagnetic transition
IC → FM

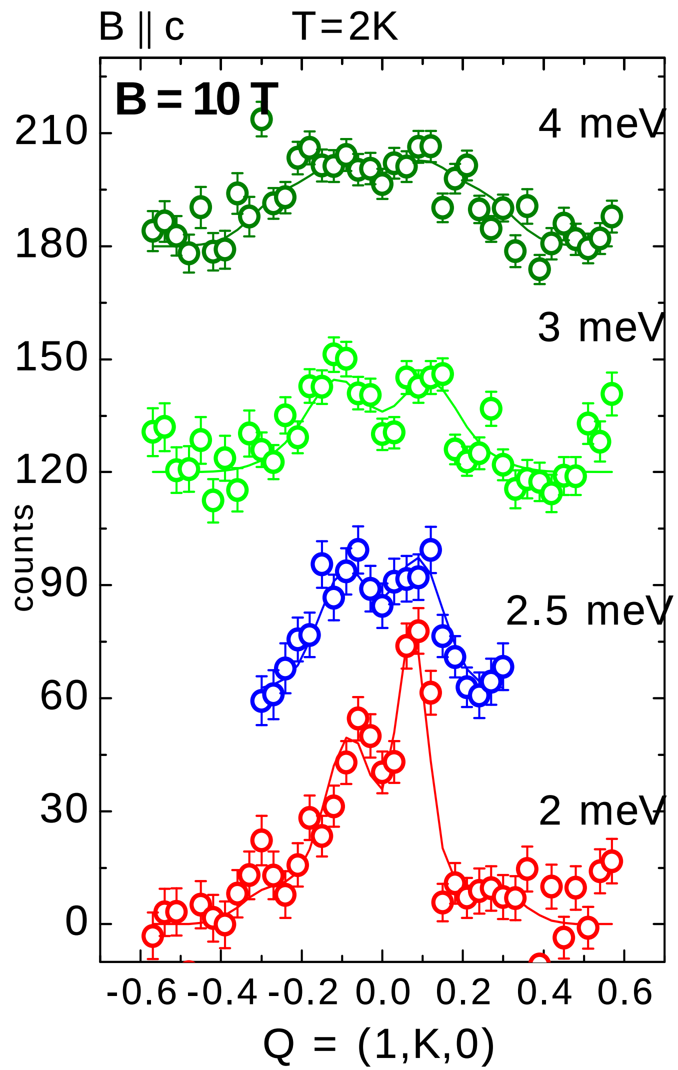


Steffens et al. Cond-mad(2010)

$Ca_{1.8}Sr_{0.2}RuO_4$: FM-magnons







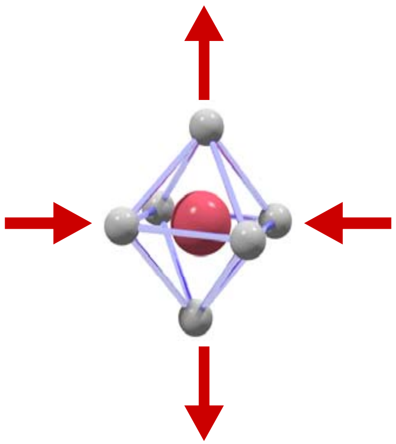
Dispersive mode (Magnon)

$\omega = D q^2$

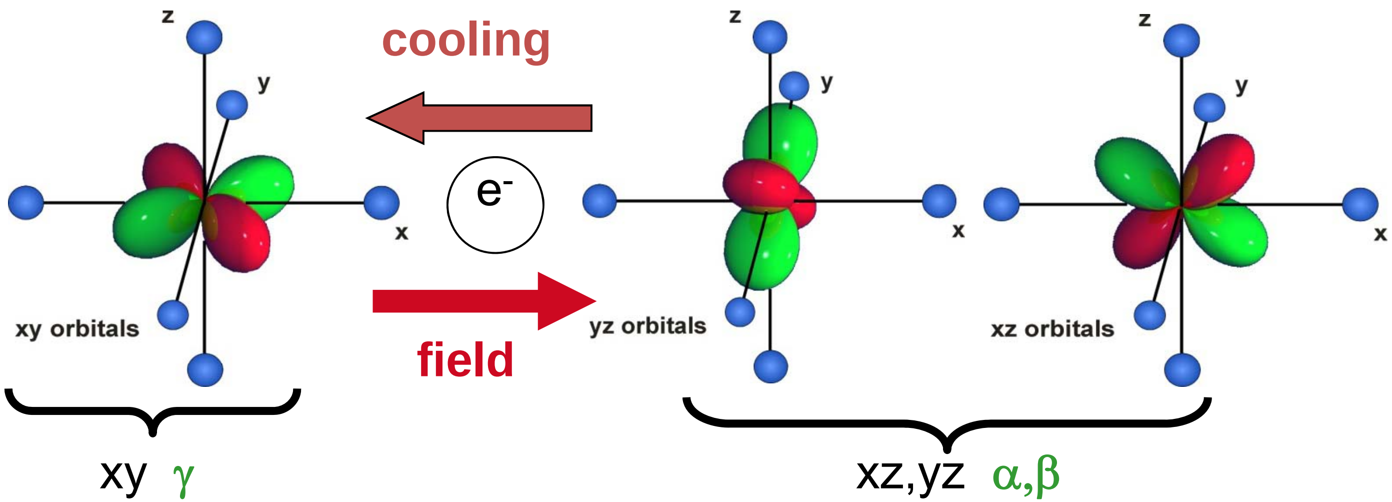
Field induced FM state

Steffens et al. Cond-mad(2010)

$Ca_{1.8}Sr_{0.2}RuO_4$: structural effects



Distortion of the octahedron



Magnetism of γ-band