IMSS Symposium, 16-17 October 2008

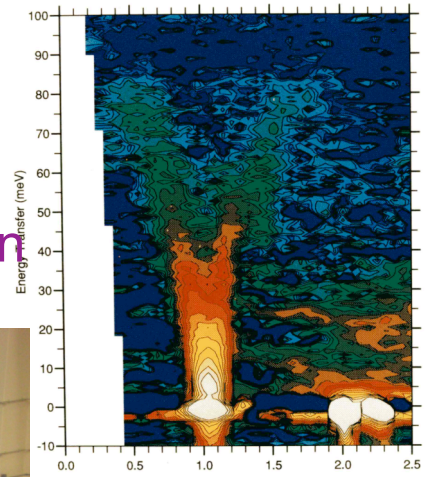
High Resolution Inelastic Neutron Scattering Studies on Spin Systems

Shinichi Itoh

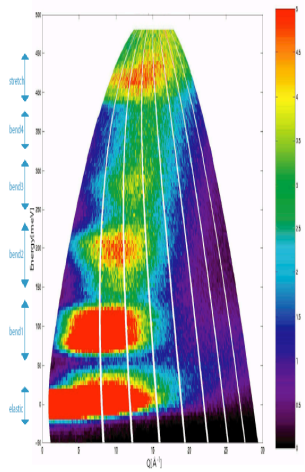
Neutron Science Division
Institute of Materials Structure Science
High Energy Accelerator Research Organization

High Resolution Chopper Spectrometer at J-PARC

This spectrometer is useful for a wide range of dynamical studies of materials with high resolutions.

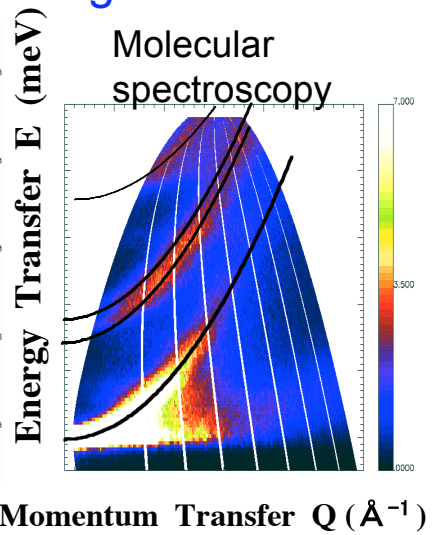


spin waves

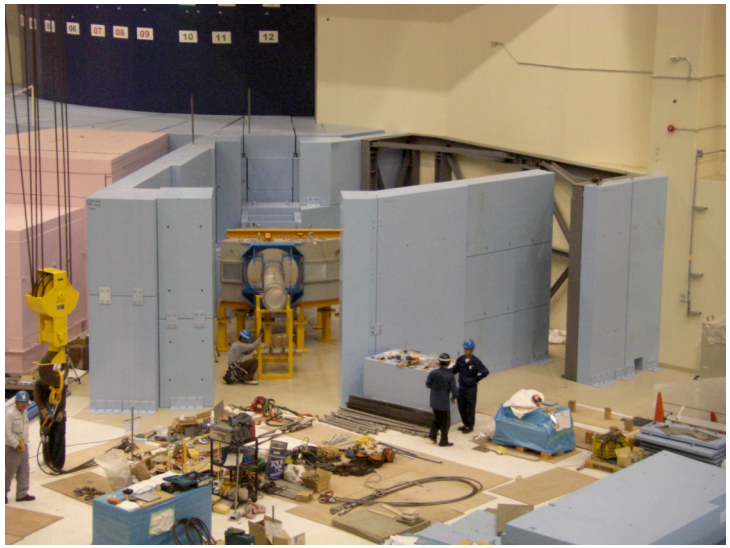


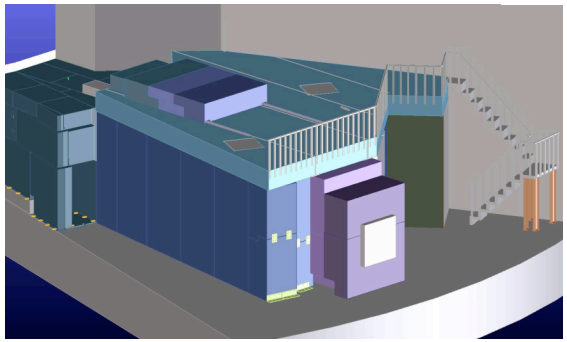
water

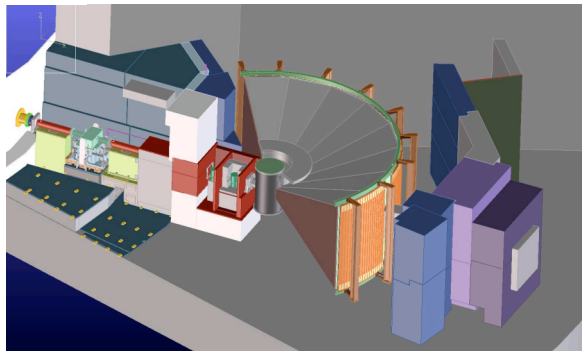
Molecular spectroscopy



Current status of construction







Dec 2008	Completion of building Shieldings
	Test experiment using KENS instrument.
Oct 2009	Experiment using full vacuum chamber + low angle dets

High Resolution Experiments ($\Delta E/E_i \sim 1\%$)
using relatively high energy neutrons (Ei ~ 0.1 - 1 eV)
for studies on spin systems

1. High resolution experiments in conventional (Q,ω) space
E_i=100meV, ΔE=1meV, ΔQ=0.03A^{-1}
Quantum phase transition and multi degree of freedom in correlated electron systems

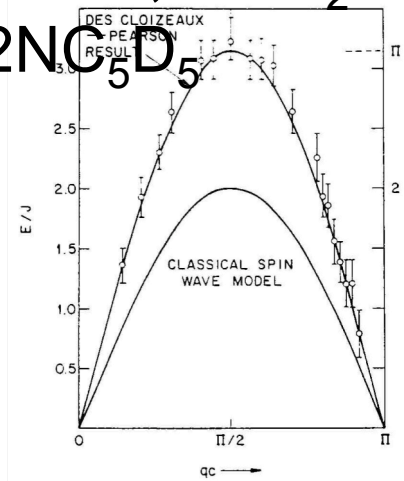
2. Access to 1st Brillouin zone with low angle and high Ei
E=10meV, Q=0.2A^{-1}
Observation of ferromagnetic spin waves from polycrystalline sample and material development

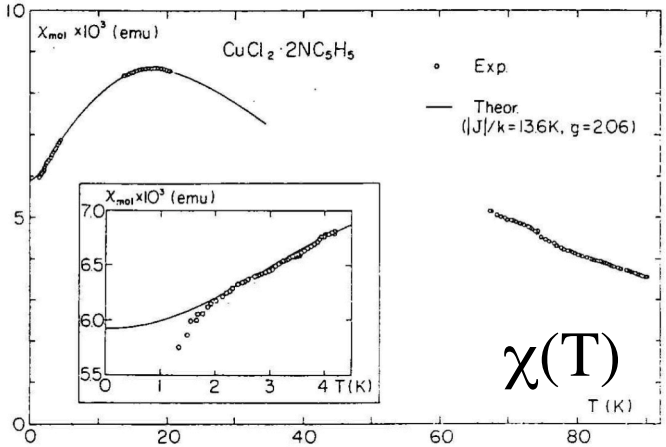
3. Possibility of eV region neutron spectroscopy
E_i=1eV, ΔE=10meV, ΔQ=0.1A^{-1}
Observations of high energy magnetic excitations
as well as electronic excitations

1. High resolution experiments in conventional (Q,ω) space

Experience: S dependent quantum renormalization in 1D AFM

S=1/2, $CuCl_2 \cdot 2NC_5D_5$





E(q)= 2πJ I sin q I

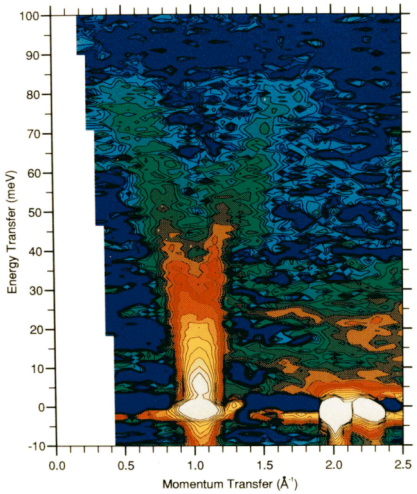
= 4SJR I sin q I

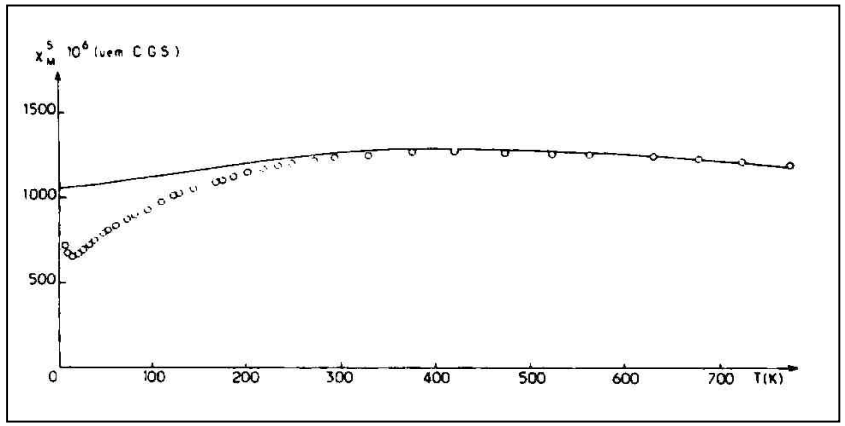
R = π/2

E(q) Y. Endoh et al. 1974

χ(T) K. Takeda et al. 1971

S=3/2,





E(q)= 4SJR I sin q I

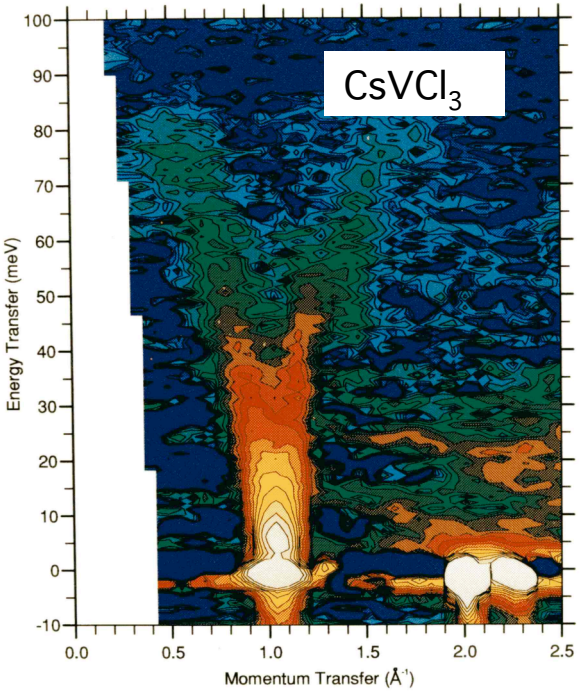
J ?, R ?

E(q) S. Itoh et al. 1995

χ(T) M. Niel et al. 1977

1. High resolution experiments in conventional (Q,ω) space

Experience: S dependent quantum renormalization in 1D AFM



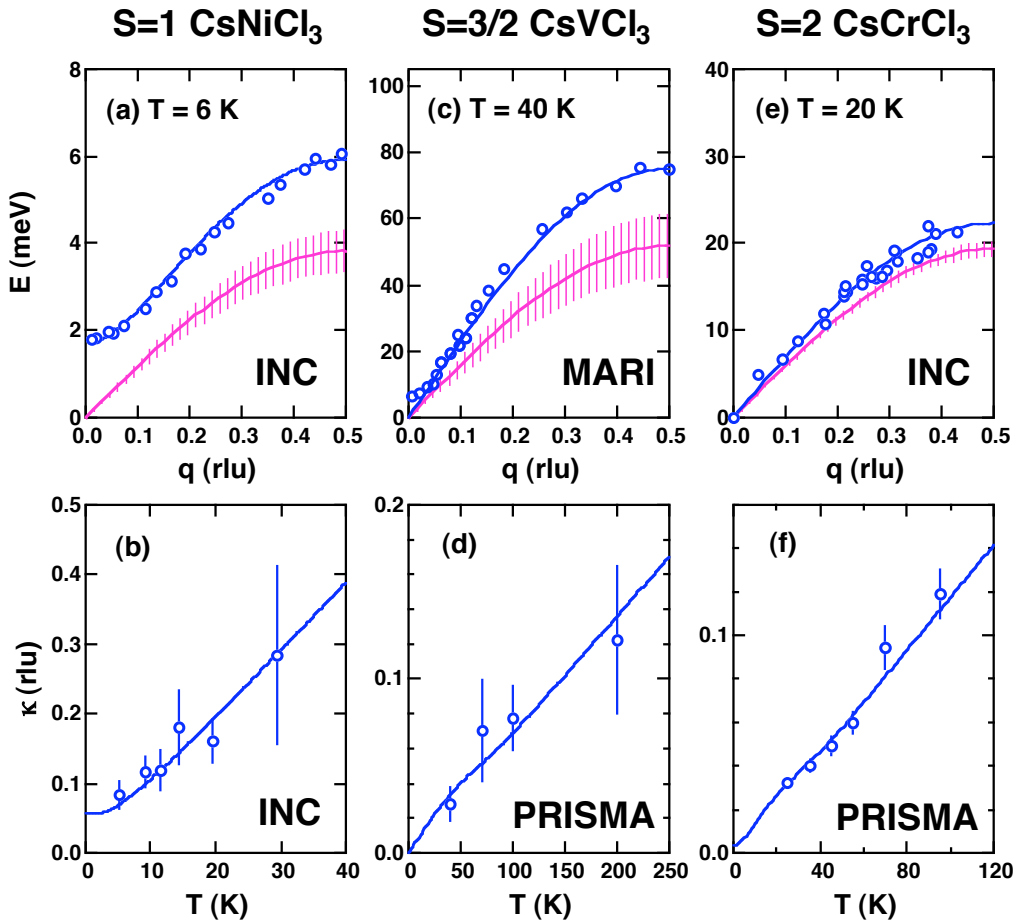
$$classical: \hbar\omega = 4SJ|\sin aq|$$

$$quantum: \hbar\omega = 4SRJ|\sin aq|$$

$$S(q,\omega) = S(q)F(q,\omega)$$

$$F(q,\omega) = \frac{\Gamma}{(\omega - E_q)^2 + \Gamma^2}$$

$$S(q) = \int S(q,\omega)d\omega = \frac{1}{q^2 + \kappa(T,J)^2}$$



S. Itoh et al., PRL 74 (1995) 2374 / JPSJ 73 (2004) 269.

E_i=125meV, $\Delta E/E_i$=3%, $\Delta q = k_i(\Delta E/E_i)/2 = 0.12$Å^{-1}

→ high resolutions are indispensable to reach the result with a single shot of experiment

1. High resolution experiments in conventional (Q,ω) space

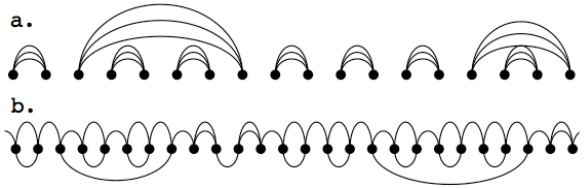
Random Singlet Phase Transition

S=1/2

By the distribution of exchange constant J, randomly selected spin pair is coupled into singlet (random singlet phase).

S=3/2

δ : Distribution of J



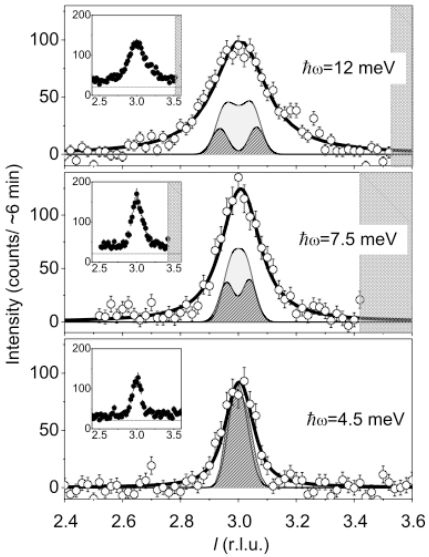
$\delta > \delta_c(S_{eff}=3/2)$

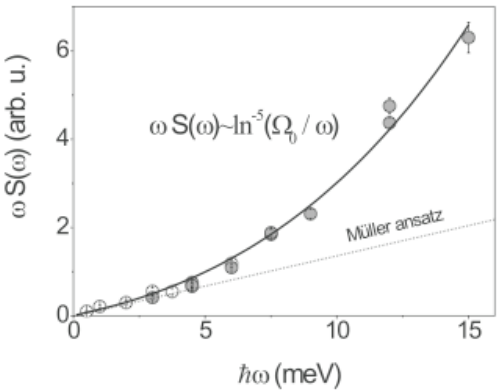
$\delta < \delta_c\ (S_{eff}=1/2)$

Phase transition between two phases at $\delta = \delta_c$.
S(q,w) shows critical exponent ψ near δ_c.

S=1/2 $BaCu_2(Si_{0.5}Ge_{0.5})_2O_7$

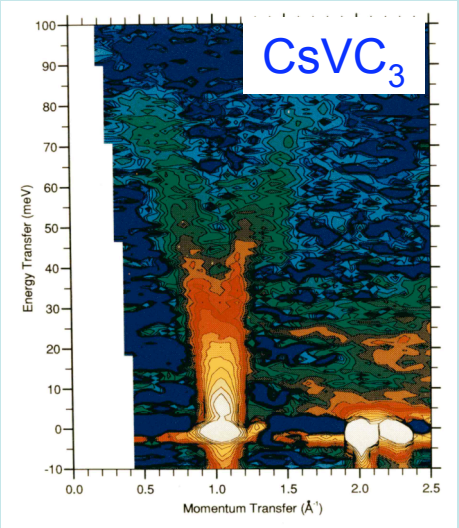
T. Masuda et al. PRL 93 (2004) 077206





$$S(q,\omega) = \frac{A}{\omega \ln^3(\Omega_0/\omega)} F\left(q \ln^2(\Omega_0/\omega)\right)$$

S=3/2 $CsV(Cl_cX_{1-c})_3$, X=Br, I



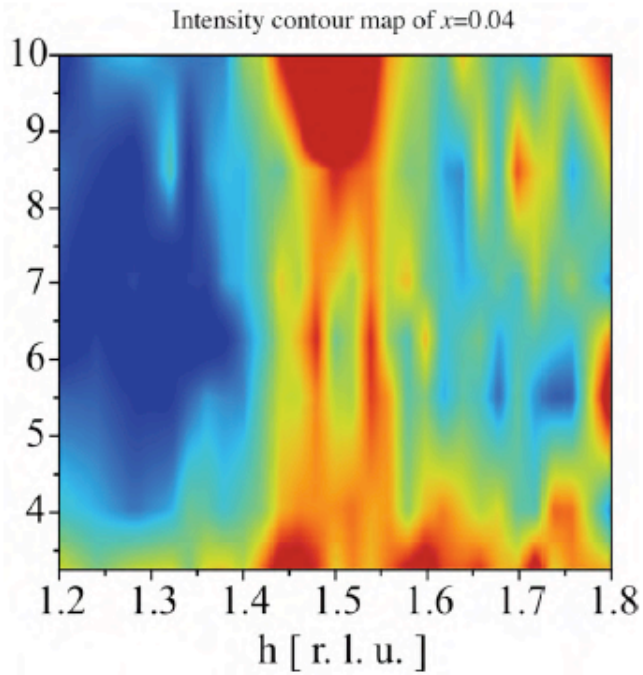
$$S(q,\omega) = \frac{A}{\omega \ln^{1+1/\psi}(\Omega_0/\omega)} \Phi\left(\left|q \ln^{1/\psi}(\Omega_0/\omega)/n_0\right|^{1/2}\right)$$

1. High resolution experiments in conventional (Q,ω) space

Dynamics of holes doped into Haldane chain

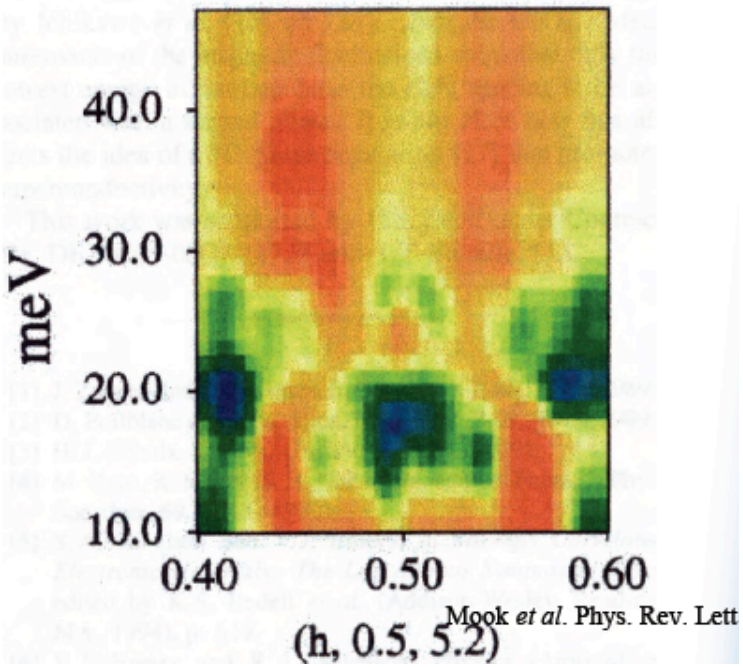
Hole-Doped 1D Heisenberg Chain

(Nd,Ca)2BaNiO5



Hole-Doped 2D High-Tc Cuprate

YBCO6.35 (Tc=39 K)



Mook *et al.* Phys. Rev. Lett

- Determination of entire dispersion relation of 1D chain
- Doping dependence of incommensurate low-E excitations

⇨ Comprehensive understanding of hole dynamics

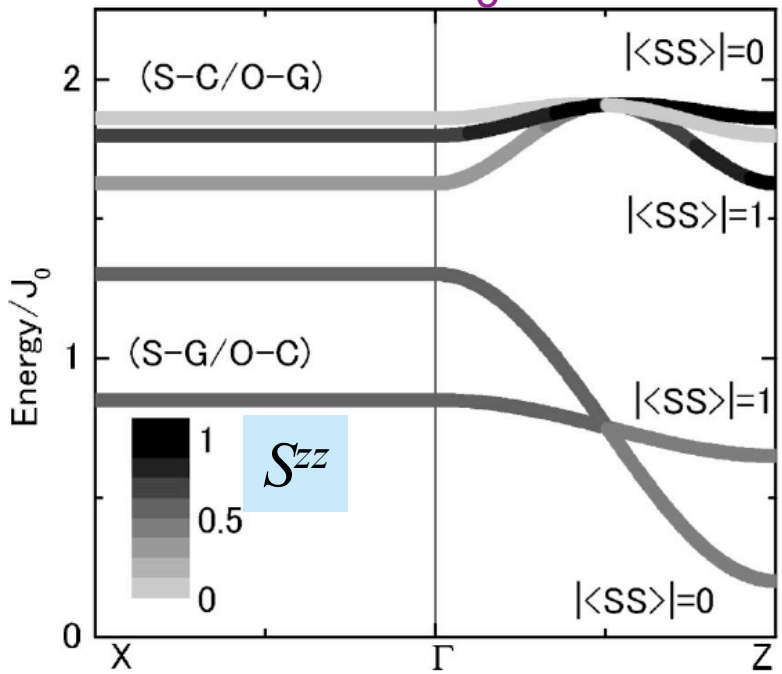
1. High resolution experiments in conventional (Q,ω) space

Possibility of observation of orbital waves in YVO_3

$$\frac{d\sigma^2}{d\Omega d\omega_f} = \left(\frac{\gamma e^2}{m_N c^2}\right)^2 \left(\frac{1}{2} g F(\vec{K})\right)^2 \frac{k_f}{k_i}$$

$$\times \sum_{l_i l_f} (\delta_{l_i l_f} - \kappa_{l_i} \kappa_{l_f}) S^{l_i l_f}(\vec{K}, \omega)$$

$$S^{l_i l_f}(\vec{r}_{id} - \vec{r}_{i'd'}, t) = \langle L^{l_i}_{id}(t) L^{l_f}_{i'd'}(0) \rangle$$



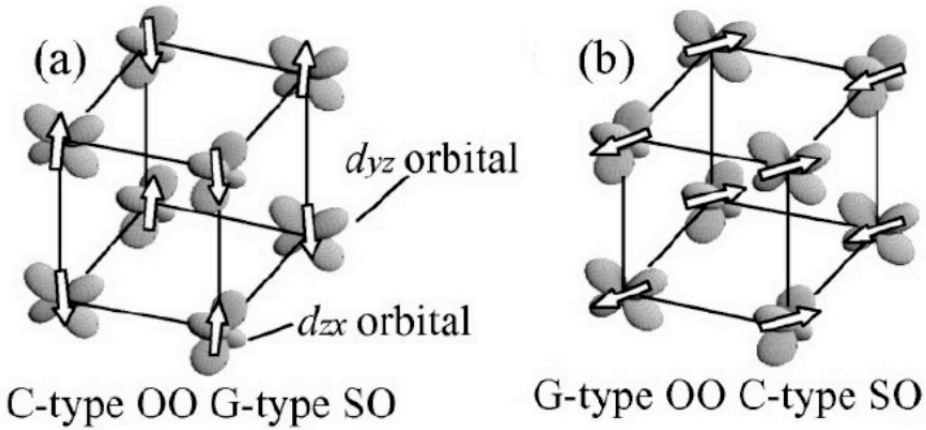
YVO_3, V^{3+} $3d^2$ t_{2g}^2

$$J_0 = 4t_0^2/(U' - I) = 33 \text{ meV}$$

4 modes

y_A ($d_{xy} \rightarrow d_{yz}$) and z_A ($d_{zx} \rightarrow d_{yz}$) at site A ($d_{xy}$ and d_{zx} occupied),

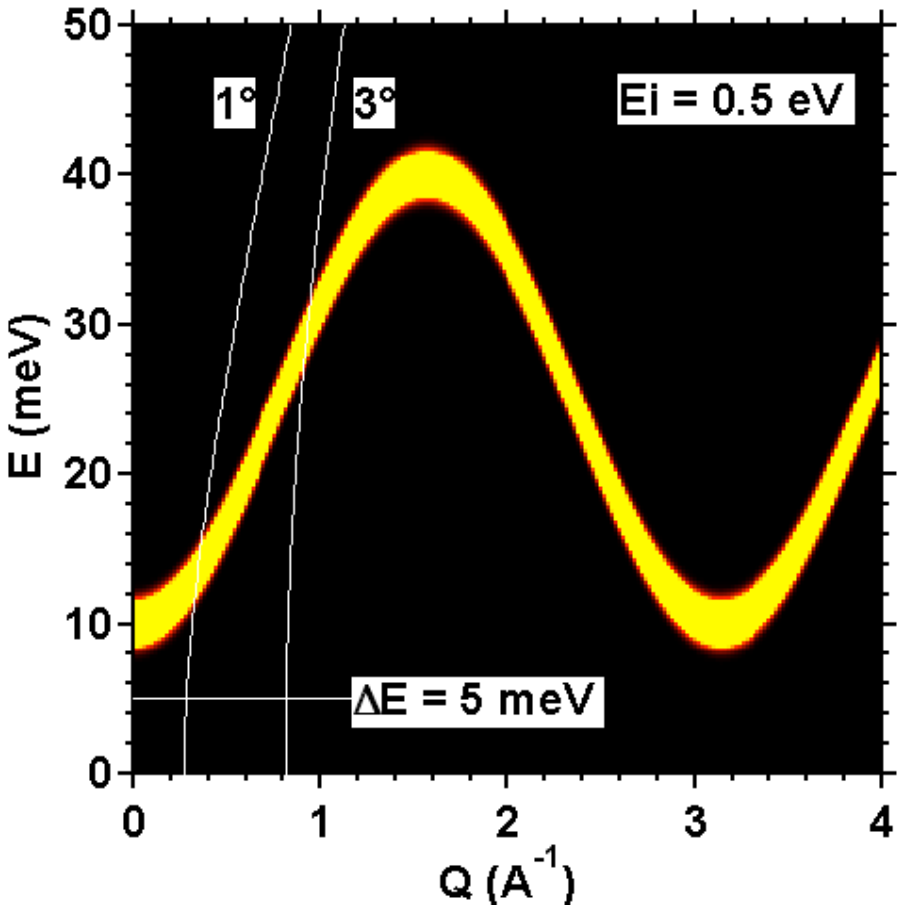
x_B ($d_{xy} \rightarrow d_{zx}$) and z_B ($d_{yz} \rightarrow d_{zx}$) at site B ($d_{xy}$ and d_{yz} occupied).



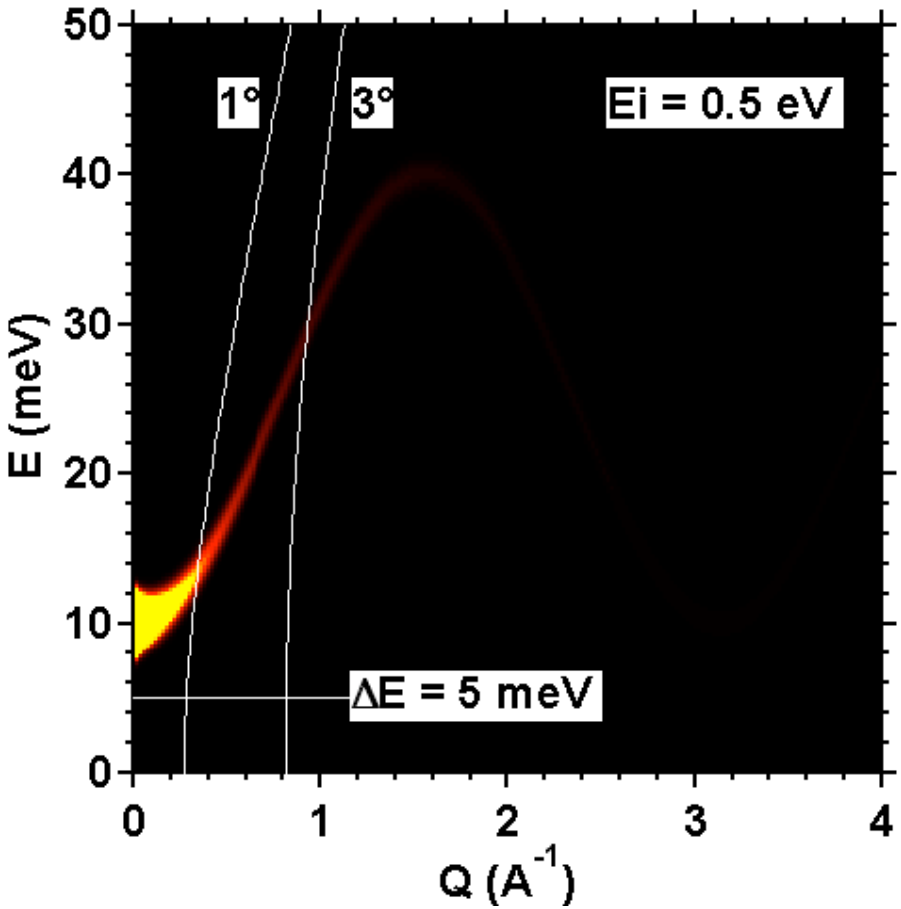
S. Ishihara: Phys. Rev. B 69 (2004) 075118, C. Ulrich *et al.*: Phys. Rev. Lett. 91(2003) 257202.

2. Access to 1st Brillouin zone with low angle and high Ei

Observation of spin waves from polycrystalline sample



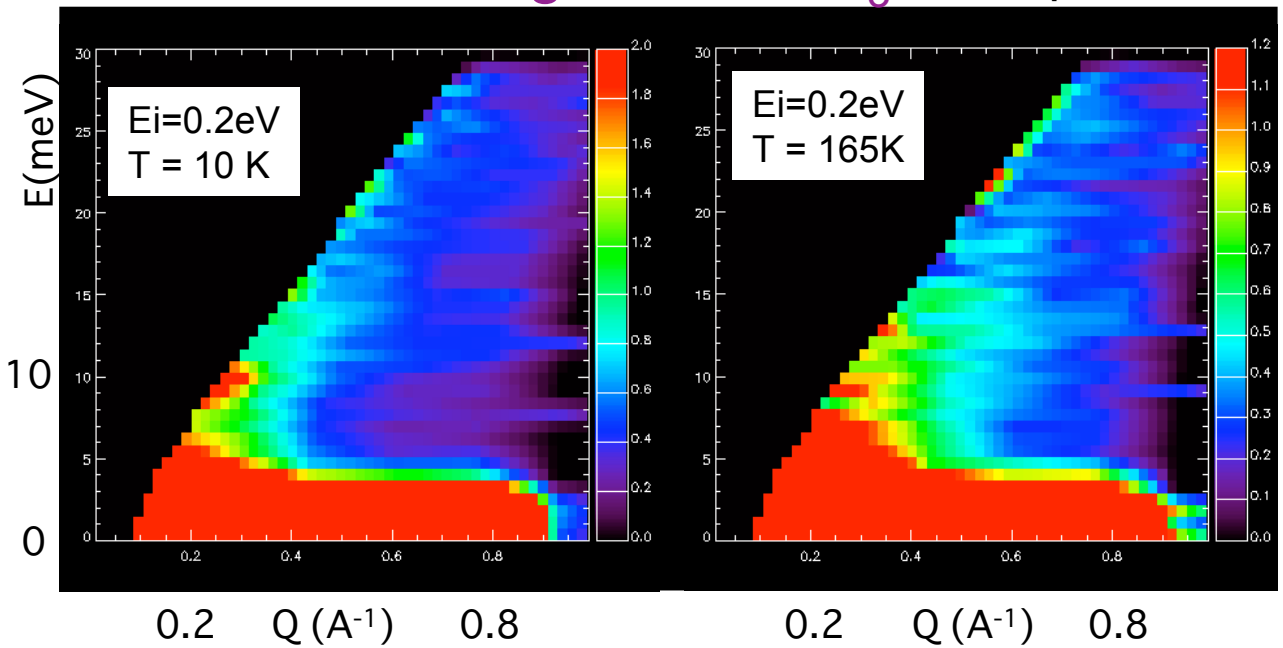
Single crystal sample

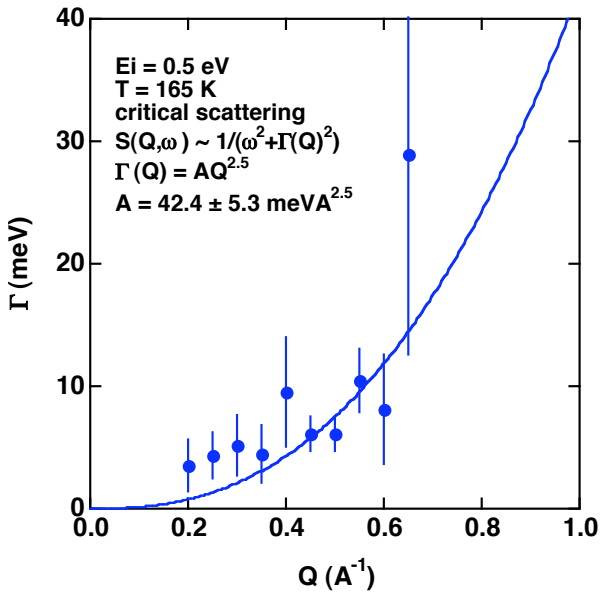


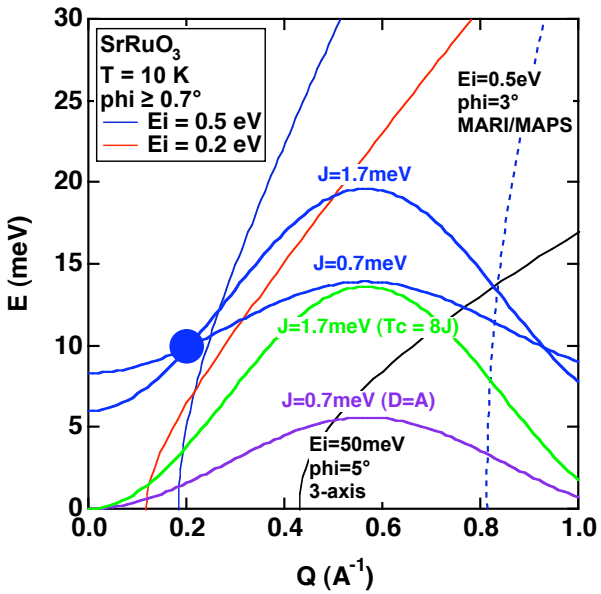
polycrystalline sample

2. Access to 1st Brillouin zone with low angle and high Ei

Metal Ferromagnet $SrRuO_3$ cubic perovskite, Tc = 160 K







Magnetic excitation at E=10meV, Q=0.2Å^{-1}
Γ(Q),Tc → J=0.7~1.7meV

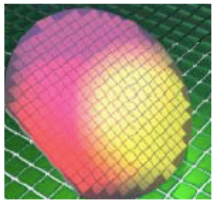
→ E_{gap} ~ several meV

High symmetry FM : E_{gap} ~ 0
Suggestion of contribution of orbitals

2. Access to 1st Brillouin zone with low angle and high Ei

Ferromagnetic Semiconductors

Semiconductor electronics



Utilize ***charge*** of electrons

Magnetic devices

Utilize ***spin*** of electrons

Spintornics --- utilize both of *charge* and *spin* of electrons

Ferromagnetic semiconductors @RT: indispensable for spintronics

GaN	+ Mn	$T_C \sim 900K$ (Sonoda)	vs	PM (Munekata)
	+ Cr	$> RT$ (Asahi)		
	+ Gd	$> RT$ (Asahi, Ploog)		
ZnO	+ Co	$T_C > RT$ (Tabata)	vs	PM (Kawasaki)
	+ Mn			
	+ Cr			
TiO_2	+ Co	$T_C > RT$ (Kawasaki)		
ZnTe	+ Cr	$T_C \sim 300K$ (Saito)		
$CdGeP_2$	+ Mn	$T_C > RT$ (Satoh)		
Ge	+ Mn	$T_C \sim 120K$ (Park)		
	+ Fe	170K (Tanaka)		

Do room temperature ferromagnetic semiconductors really exist?
What is the mechanism of ferromagnetism, if the materials exist?

2. Access to 1st Brillouin zone with low angle and high Ei

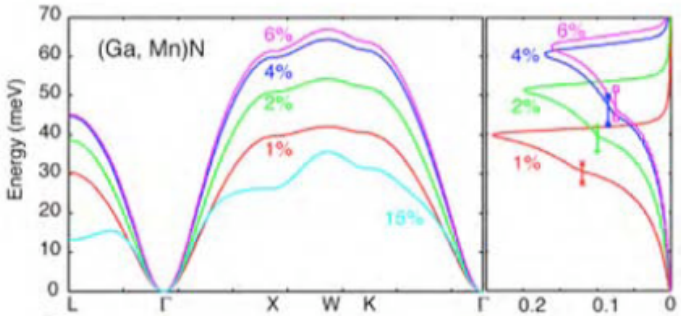
Ferromagnetic Semiconductors

Possible mechanisms of ferromagnetism in semiconductors

1. carrier induced interaction

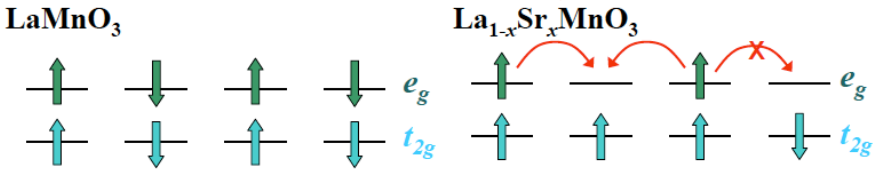
high carrier density: FM low carrier density: PM

Carriers (holes) mediates FM interaction.



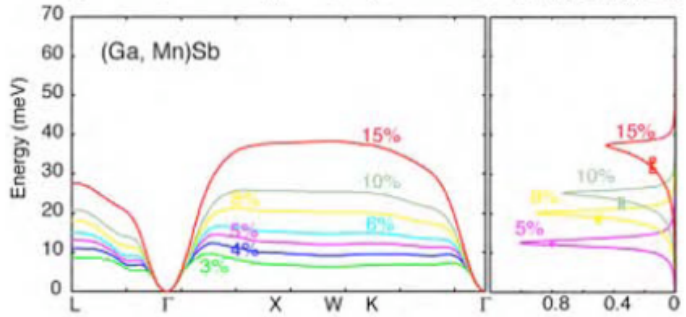
Short range interaction: dispersive spin waves

2. double exchange interaction



insulator:AFM

parallel spins hopping by hole-doping:

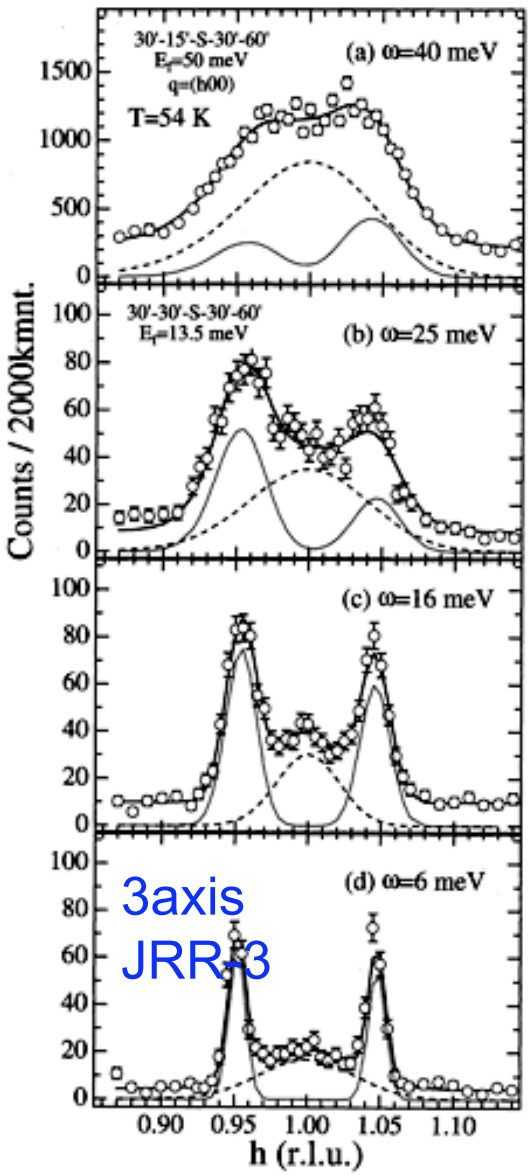


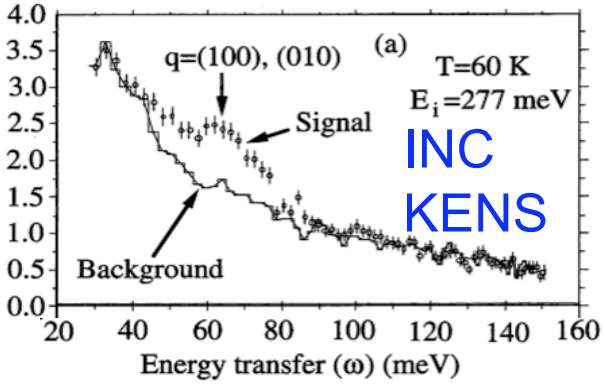
long range interaction: flat spin waves

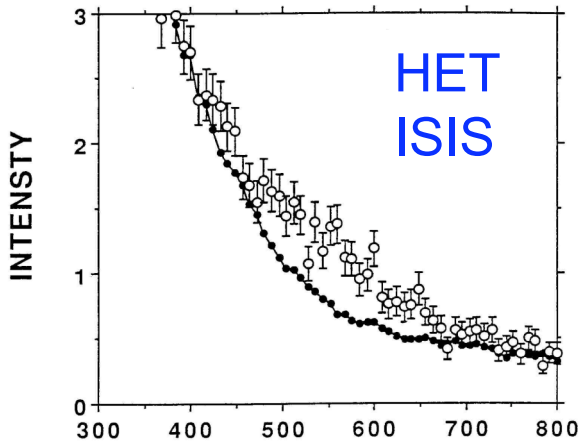
Determination of mechanism by measuring spin wave dispersion
Almost samples are thin films. We are looking for bulk samples for demonstrations of inelastic neutron experiments.

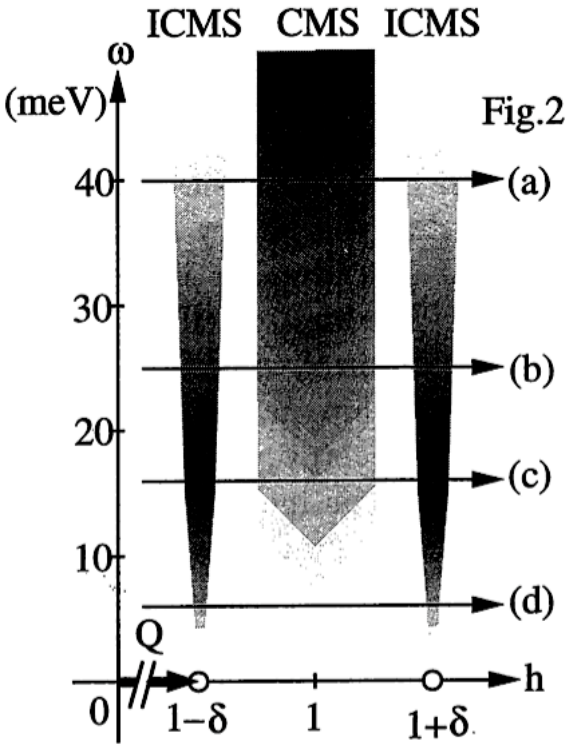
3. Possibility of eV region neutron spectroscopy

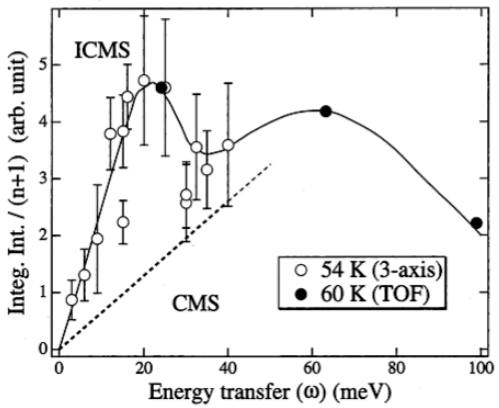
High Energy Magnetic Excitations in Cr





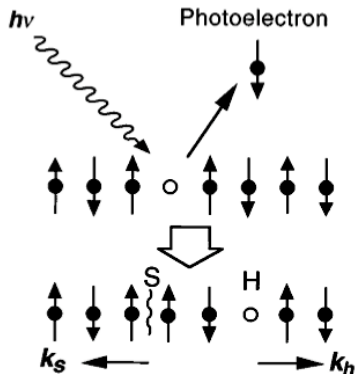


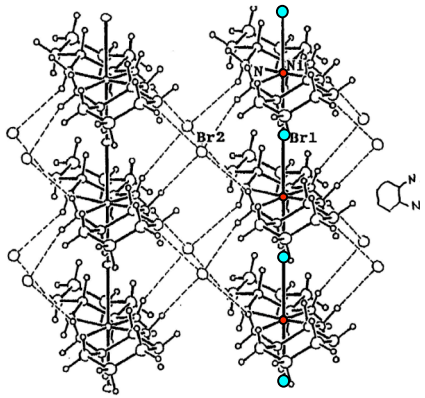




3. Possibility of eV region neutron spectroscopy

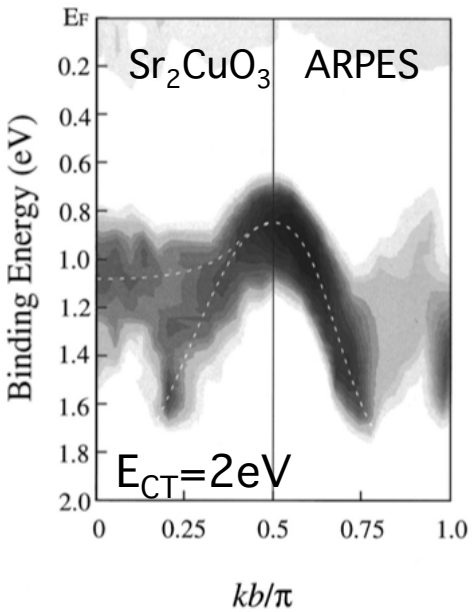
Electronic excitations

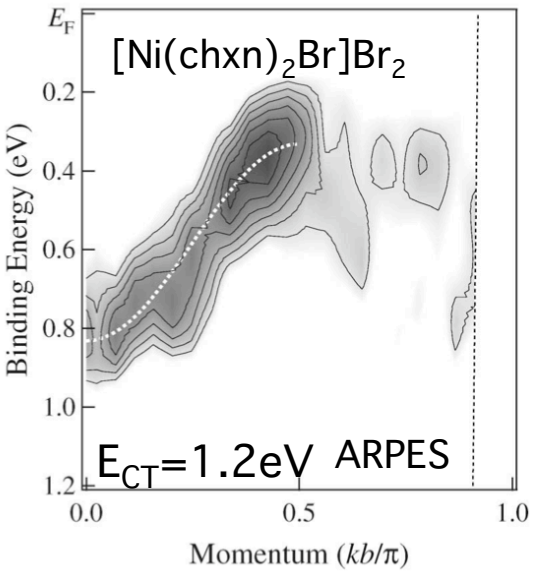




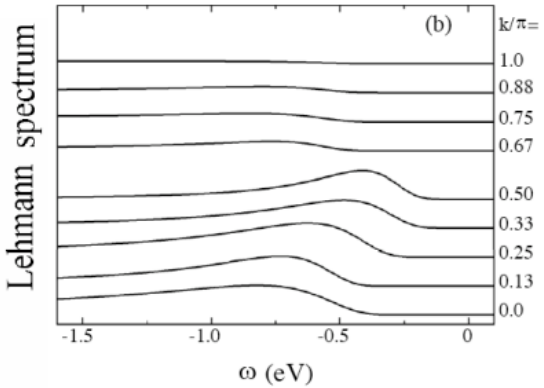
$$H = -t\sum_l (a^+_{l\sigma}a_{l+1\sigma} + a^+_{l+1\sigma}a_{l\sigma}) + U\sum_l n_{l\uparrow}n_{l\downarrow} + V\sum_l n_l n_{l+1}$$

The dipole transition in the 1 -D system. The white circle denotes the hole site, while the black circle denotes the doubly occupied site.





E_{CT} decreases via perturbation of V(inter-site Coulomb).



The theoretical calculation well agrees with the result of ARPES.

Clear spin-charge separation

unclear spin-charge separation

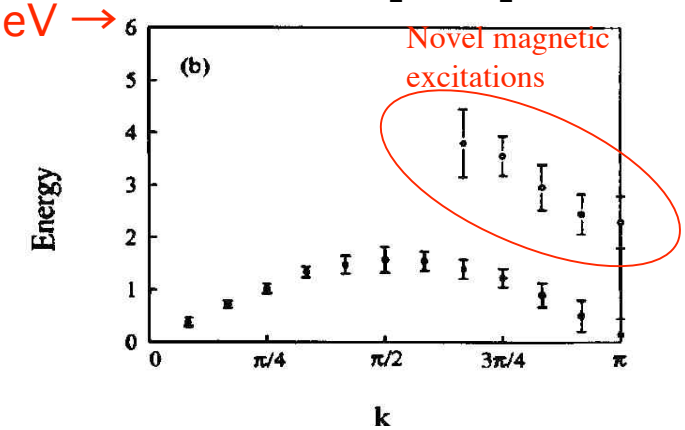
Sr_2CuO_3	H. Fujisawa et al., PRB 59 (1999) 7358.
$SrCuO_2$	C. Kim et al., PRL 77 (1996) 4054.
$[Ni(chxn)_2Br]Br_2$	S. Fujimori et al., PRL 88 (2002) 247601.

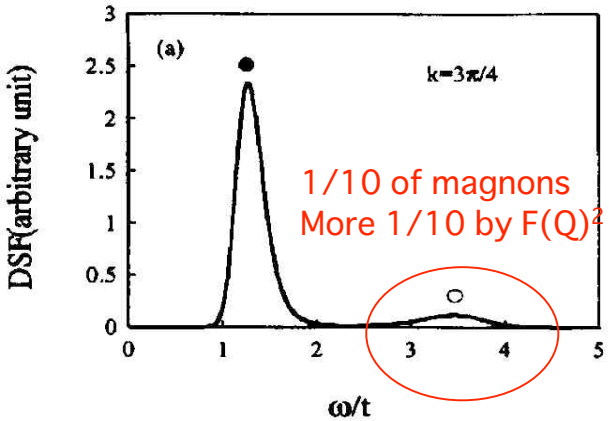
3. Possibility of eV region neutron spectroscopy

Electronic excitations

$[Ni(chxn)_2Br]Br_2$

1eV →





$$H = -t\sum_l (a^+_{l\sigma}a_{l+1\sigma} + a^+_{l+1\sigma}a_{l\sigma}) + U\sum_l n_{l\uparrow}n_{l\downarrow} + V\sum_l n_l n_{l+1}$$

1D extend Hubberd model

N. Tomita and K. Nasu, Phys. Rev. B 61 (2000) 2488

Magnetic excitation energies become the same order as charge excitation energies via inter-site Coulomb repulsion.

Spin-induced charge excitations can be observed as magnetic scattering.

J-PARC 1MW

E_i = 1 eV, $\Delta E/E_i$ = 1%

N = 100, $\Delta N/N$ = 10%

400 g days = 40g x 10days

similar experiment to current high-Tc studies at ISIS

High Resolution Experiments ($\Delta E/E_i \sim 1\%$)
using relatively high energy neutrons (Ei ~ 0.1 - 1 eV)
for studies on spin systems

1. High resolution experiments in conventional (Q,ω) space
E_i=100meV, ΔE=1meV, ΔQ=0.03A^{-1}
Quantum phase transition and multi degree of freedom in correlated electron systems

2. Access to 1st Brillouin zone with low angle and high Ei
E=10meV, Q=0.2A^{-1}
Observation of ferromagnetic spin waves from polycrystalline sample and material development

3. Possibility of eV region neutron spectroscopy
E_i=1eV, ΔE=10meV, ΔQ=0.1A^{-1}
Observations of high energy magnetic excitations
as well as electronic excitations