High Resolution Inelastic Neutron Scattering Studies on Spin Systems

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High Resolution Chopper Spectrometer at J-PARC

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

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 ($E_i \sim 0.1 - 1$ eV) for studies on spin systems

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

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

3. Possibility of eV region neutron spectroscopy $E_i=1$ eV, $\Delta E=10$ meV, $\Delta Q=0.1 A^{-1}$
Observations of high energy magnetic excitations as well as electronic excitations
1. High resolution experiments in conventional $(Q, \omega)$ space

Experience: $S$ dependent quantum renormalization in 1D AFM

$S=1/2$, $\text{CuCl}_2 \cdot 2N\text{C}_5\text{D}_5$

$E(q) = 2\pi J | \sin q |$

$I = 4SJ\pi | \sin q |$

$R = \pi/2$

$E(q)$ Y. Endoh et al. 1974

$\chi(T)$ K. Takeda et al. 1971

$S=3/2$, $\text{CsVCl}_3$

$E(q) = 4S\pi J | \sin q |$

$J \ ?, R \ ?$

$E(q)$ S. Itoh et al. 1995

$\chi(T)$ M. Niel et al. 1977
1. High resolution experiments in conventional \((Q, \omega)\) space

Experience: \(S\) dependent quantum renormalization in 1D AFM

\[
S(q, \omega) = S(q)F(q, \omega)
\]

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

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

\(S_1\) \(\text{CsNiCl}_3\) \(S_{3/2}\) \(\text{CsVCl}_3\) \(S_2\) \(\text{CsCrCl}_3\)

(a) \(T = 6\ \text{K}\) \(\quad\) (c) \(T = 40\ \text{K}\) \(\quad\) (e) \(T = 20\ \text{K}\)

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

\[\rightarrow\] high resolutions are indispensable to reach the result with a single shot of experiment
1. High resolution experiments in conventional $(Q, \omega)$ 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).

$S=1/2 \quad \text{BaCu}_2(\text{Si}_{0.5}\text{Ge}_{0.5})_2\text{O}_7$

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

$S=3/2 \quad \text{CsV(Cl}_{c}\text{X}_{1-c})_3$, X=Br, I

$S=3/2$

$\delta$ : Distribution of $J$

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

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

Phase transition between two phases at $\delta = \delta_c$.

$S(q,\omega)$ shows critical exponent $\psi$ near $\delta_c$.

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

$\phi$ : Distribution of $J$

$\phi > \phi_c (S_{\text{eff}}=3/2)$

$\phi < \phi_c (S_{\text{eff}}=1/2)$

CsVC$_3$
1. High resolution experiments in conventional \((Q, \omega)\) space
Dynamics of holes doped into Haldane chain

- 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 \((Q, \omega)\) space

Possibility of observation of orbital waves in \(\text{YVO}_3\)

\[
\frac{d\sigma^2}{d\Omega d\omega_f} = \left( \frac{\gamma e^2}{m_N c^2} \right)^2 \left( \frac{1}{2} g F(\vec{K}) \right)^2 \frac{k_f}{k_i} \times \sum_{l_i l_f} (\delta_{l_i l_f} - \kappa_{l_i} \kappa_{l_f}) S^{l_i l_f}(\vec{K}, \omega)
\]

\[
S^{l_i l_f}(\vec{r}_{i d} - \vec{r}_{i' d'}, t) = \langle L_{i d}^{l_i}(t) L_{i' d'}^{l_f}(0) \rangle
\]

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

\(\text{YVO}_3, \text{V}^{3+} 3d^2 t_{2g}^2\)

4 modes

\(y_A (d_{xy} \rightarrow d_{yz})\) and \(z_A (d_{zx} \rightarrow d_{yx})\)

at site A \((d_{xy} \text{ and } d_{zx}\) occupied),

\(x_B (d_{xy} \rightarrow d_{zx})\) and \(z_B (d_{yz} \rightarrow d_{zx})\)

at site B \((d_{xy} \text{ and } d_{yz}\) occupied).

2. Access to 1st Brillouin zone with low angle and high $E_i$
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 $E=10\text{meV}$, $Q=0.2\text{Å}^{-1}$

$\Gamma(Q), T_c \rightarrow J = 0.7 \sim 1.7\text{meV}$

$E_{\text{gap}} \sim \text{several meV}$

High symmetry FM : $E_{\text{gap}} \sim 0$

Suggestion of contribution of orbitals
2. Access to 1st Brillouin zone with low angle and high $E_i$

Ferromagnetic Semiconductors

**Ferromagnetic semiconductors @RT: indispensable for spintronics**

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**Semiconductor electronics**

Utilize **charge** of electrons

**Magnetic devices**

Utilize **spin** of electrons

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

Ferromagnetic semiconductors @RT: indispensable for spintronics

<table>
<thead>
<tr>
<th>Material</th>
<th>Additives</th>
<th>$T_c$ (K)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>GaN</td>
<td>Mn</td>
<td>$T_c \sim 900$</td>
<td>(Sonoda) vs PM (Munekata)</td>
</tr>
<tr>
<td></td>
<td>Cr</td>
<td>$&gt; RT$</td>
<td>(Asahi)</td>
</tr>
<tr>
<td></td>
<td>Gd</td>
<td>$&gt; RT$</td>
<td>(Asahi, Ploog)</td>
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<tr>
<td>ZnO</td>
<td>Co</td>
<td>$T_c &gt; RT$</td>
<td>(Tabata) vs PM (Kawasaki)</td>
</tr>
<tr>
<td></td>
<td>Mn</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Cr</td>
<td></td>
<td></td>
</tr>
<tr>
<td>TiO$_2$</td>
<td>Co</td>
<td>$T_c &gt; RT$</td>
<td>(Kawasaki)</td>
</tr>
<tr>
<td>ZnTe</td>
<td>Cr</td>
<td>$T_c \sim 300$</td>
<td>(Saito)</td>
</tr>
<tr>
<td>CdGeP$_2$</td>
<td>Mn</td>
<td>$T_c &gt; RT$</td>
<td>(Satohi)</td>
</tr>
<tr>
<td>Ge</td>
<td>Mn</td>
<td>$T_c \sim 120$</td>
<td>(Park)</td>
</tr>
<tr>
<td></td>
<td>Fe</td>
<td>170K</td>
<td>(Tanaka)</td>
</tr>
</tbody>
</table>

**Do room temperature ferromagnetic semiconductors really exist?**

**What is the mechanism of ferromagnetism, if the materials exist?**
Possible mechanisms of ferromagnetism in semiconductors

1. carrier induced interaction
   - high carrier density: FM
   - low carrier density: PM
   - Carriers (holes) mediate FM interaction.

2. double exchange interaction
   - insulator: AFM
   - parallel spins hopping by hole-doping:

Short range interaction: dispersive spin waves
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

INC
KENS

HET
ISIS

3axis
JRR-3

ICMS
CMS
ICMS

Fig.2

(a)
(b)
(c)
(d)

ICMS
CMS
ICMS

Integ. Int. / (μE) (arb. unit)

Q

1-δ
1
1+δ

h

Energy transfer (ω) (meV)

0

20

40

60

80

100

120

140

160

Energy transfer (ω) (meV)

0

20

40

60

80

100

Counts / 2000 kmnt.

T=54 K

q=(100), (010)

T=60 K

E_i=277 meV

E_i=13.5 meV

E_i=50 meV

ω=40 meV

ω=25 meV

ω=16 meV

ω=6 meV
3. Possibility of eV region neutron spectroscopy

Electronic excitations

\[ H = -t \sum_i (a_{i+1}^+ a_{i+1} + a_{i+1} a_{i+1}^+ a_{i+1}) + U \sum_i n_i n_{i+1} + V \sum_i n_i n_{i+1} \]

The theoretical calculation well agrees with the result of ARPES.

E\text{\textsubscript{CT}} decreases via perturbation of V\text{\textsubscript{inter-site Coulomb}}.

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

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Sr\textsubscript{2}CuO\textsubscript{3}  
SrCuO\textsubscript{2}  
[Ni(chxn)\textsubscript{2}Br]Br\textsubscript{2}  
H. Fujisawa et al., PRB 59 (1999) 7358.  
C. Kim et al., PRL 77 (1996) 4054.  
3. Possibility of eV region neutron spectroscopy

Electronic excitations

\[ [\text{Ni(chxn)}_2 \text{Br}] \text{Br}_2 \]

1 eV →

Novel magnetic excitations

\[ H = -i \sum_l (a_{l+1}^+ a_{l+1\sigma} + a_{l+1\sigma} a_{l\sigma}) + U \sum_l n_{l\uparrow} n_{l\downarrow} + V \sum_l n_{l\uparrow} n_{l+1\uparrow} \]

1D extend Hubbard model


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

\[ E_i = 1 \text{ eV}, \Delta E/E_i = 1\% \]

\[ N = 100, \Delta N/N = 10\% \]

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 \( (E_i \sim 0.1 - 1 \text{ eV}) \)
for studies on spin systems

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