



**SMR.959 - 37**

**MINIWORKSHOP ON STRONG ELECTRON CORRELATIONS**  
**"Disorder and Interaction in Quantum Systems**  
**and Their Classical Analogs"**

**(1 - 19 July 1996)**

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**"Magnetic Properties of Low Dimensional**  
**Quantum Spin Systems  $\text{CaV}_n\text{O}_{2n+1}$  ( $n=2-4$ )"**

**Masatoshi Sato**  
**Nagoya University**  
**Department of Physics**  
**Nagoya 464-01**  
**Japan**

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***These are preliminary lecture notes, intended only for distribution to participants.***

# Magnetic Properties of Low Dimensional Quantum Spin Systems $\text{CaV}_n\text{O}_{2n+1}$ ( $n=2-4$ )

Masatoshi SATO

Dept. of Phys. Nagoya Univ.

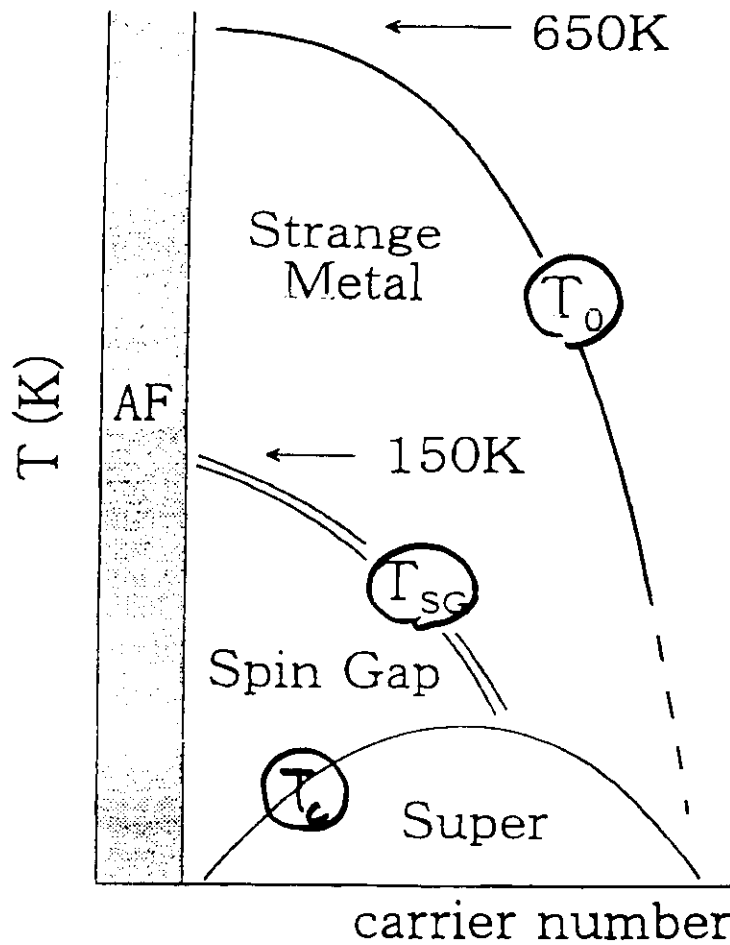
- Brief survey of anomalous metallic phase of Cu-oxides
- Experimental results of spin gap phase of  $\text{CaV}_4\text{O}_9$

Co-workers

Nagoya Univ. Group.

ISSP Group

# Anomalous Metallic Phase of High- $T_c$ Oxides

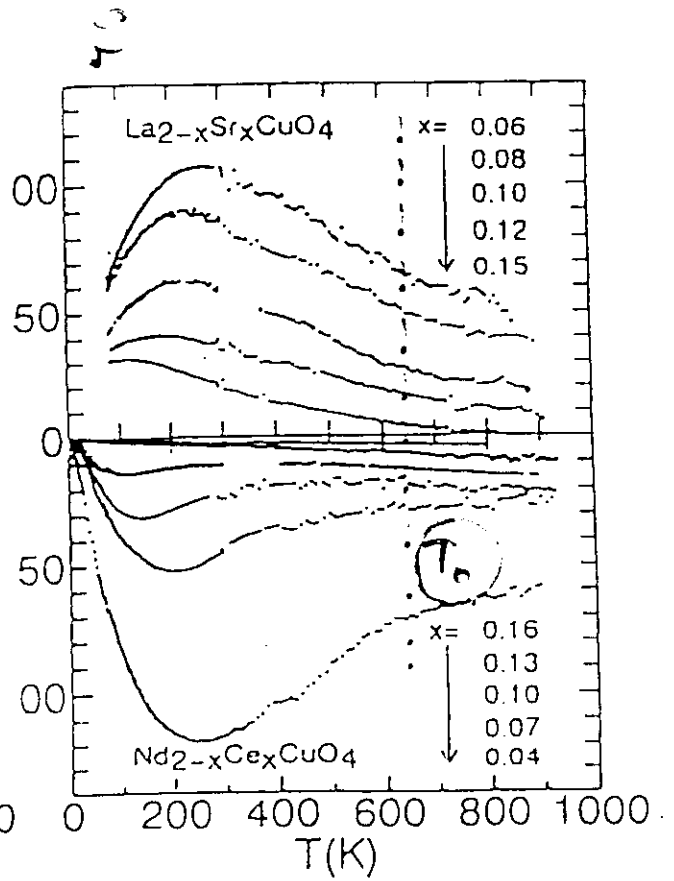
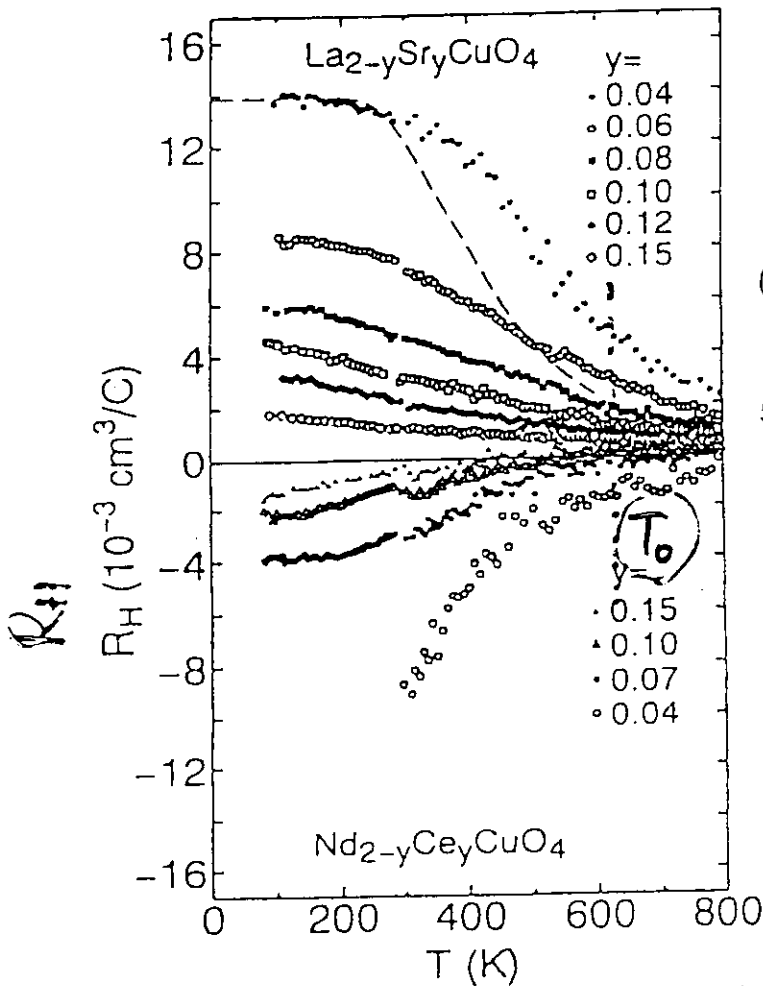
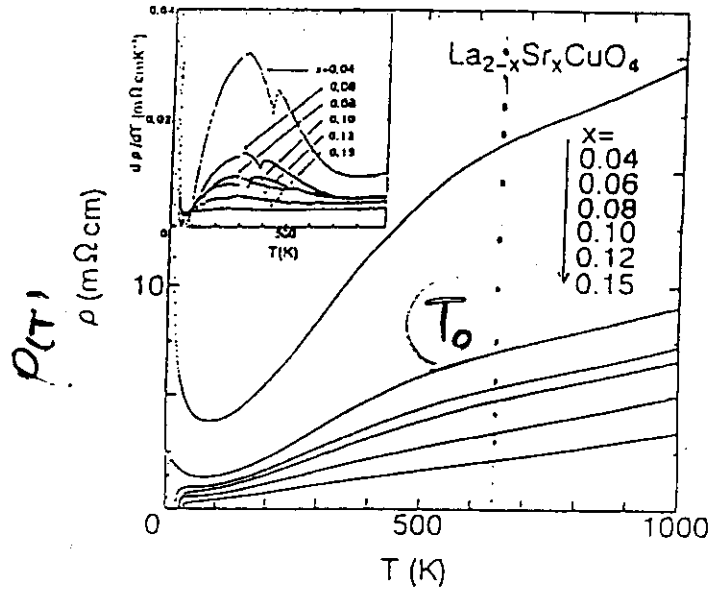


← 2D system near the Mott transition

# Anomalous Properties in Cu-Oxides

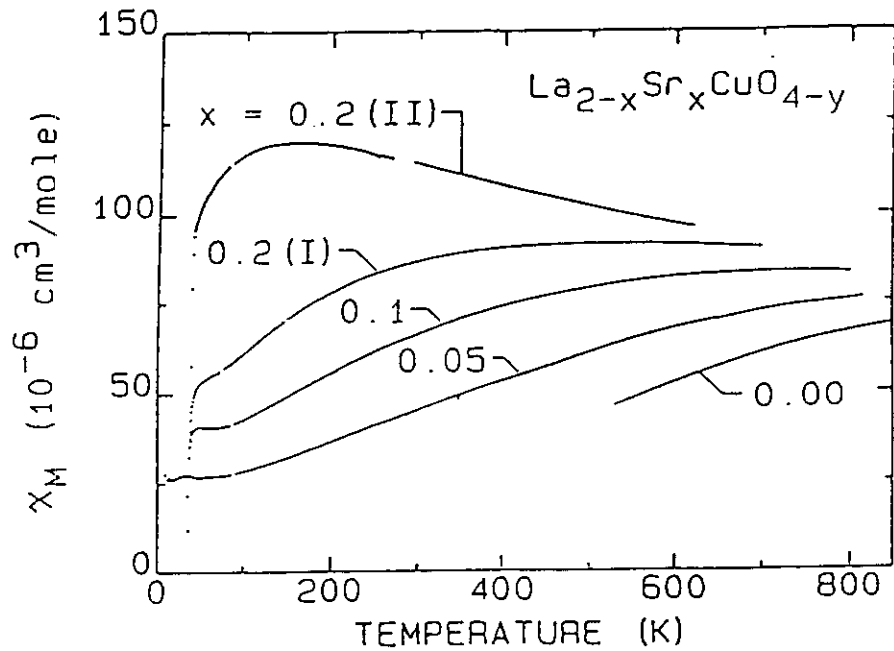
## (I) Transport Anomalies at $T \sim T_0$ ( $> 500\text{K}$ )

( $\&$  in  $x$  and  $K$ )



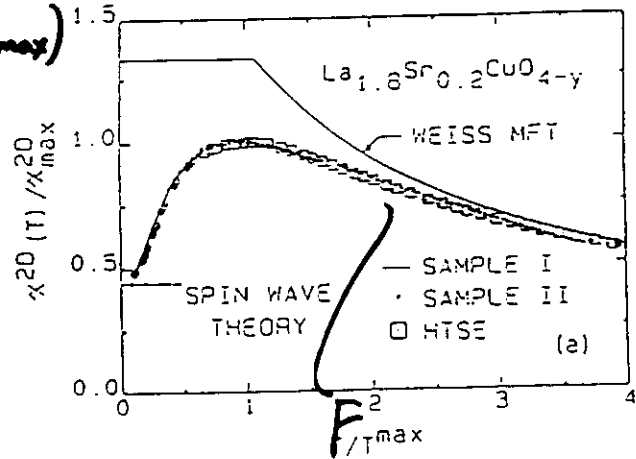
Nishikawa, Takeda, M. S.

# magnetic susceptibility $\chi$



$$\chi(T) = \chi_0 + (\chi_{\text{max}} - \chi_0) F(T/T_{\text{max}})$$

D.C. Johnston

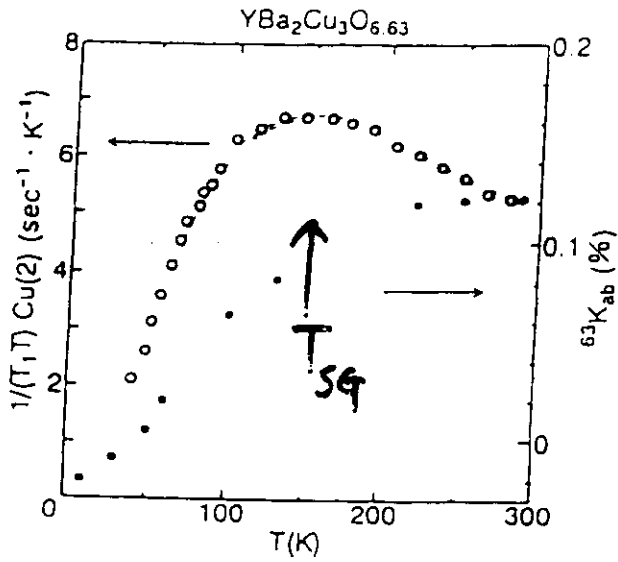


$$T_{\text{max}} \sim T_0$$

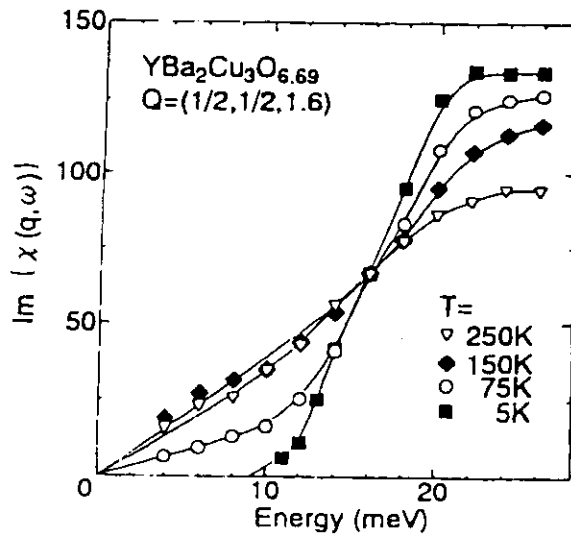
T. Nakano et al.

(R. Yoshizaki et al.)

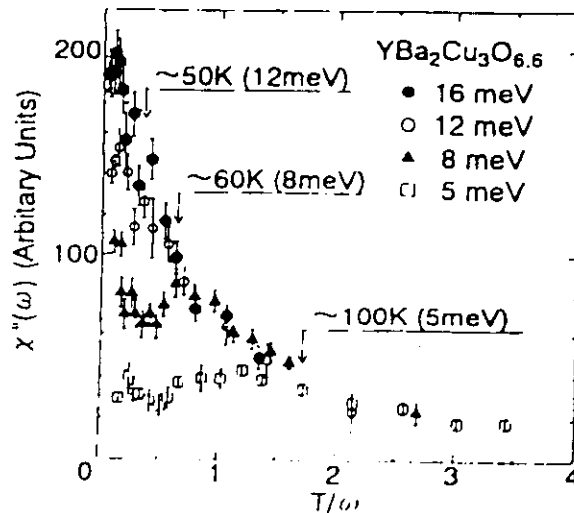
# Spin gap



NMR  $1/T_1$   
(Yasuda, Takigawa)

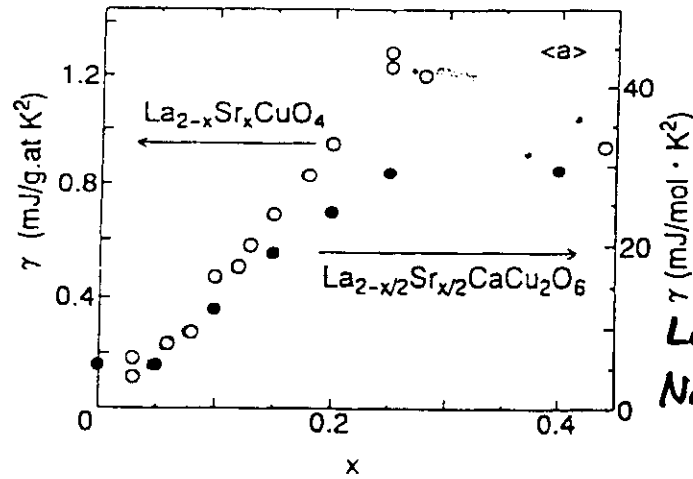


Neutron  $\chi''(\omega)$   
(Rosset-Mignod et al)

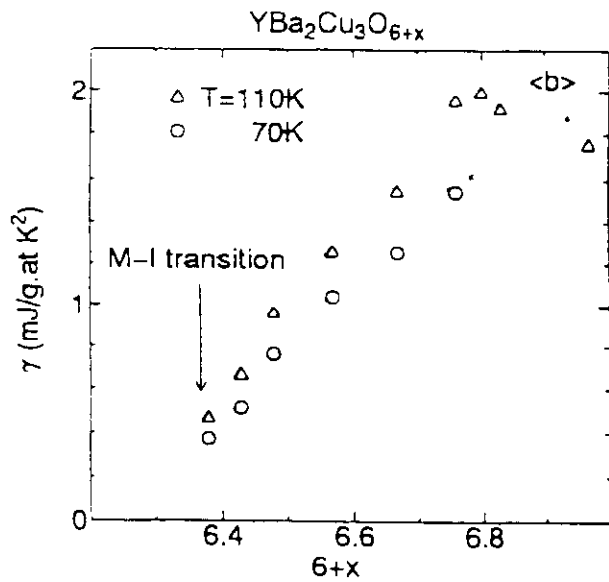


Our data

# Anomalous behavior of $\gamma$



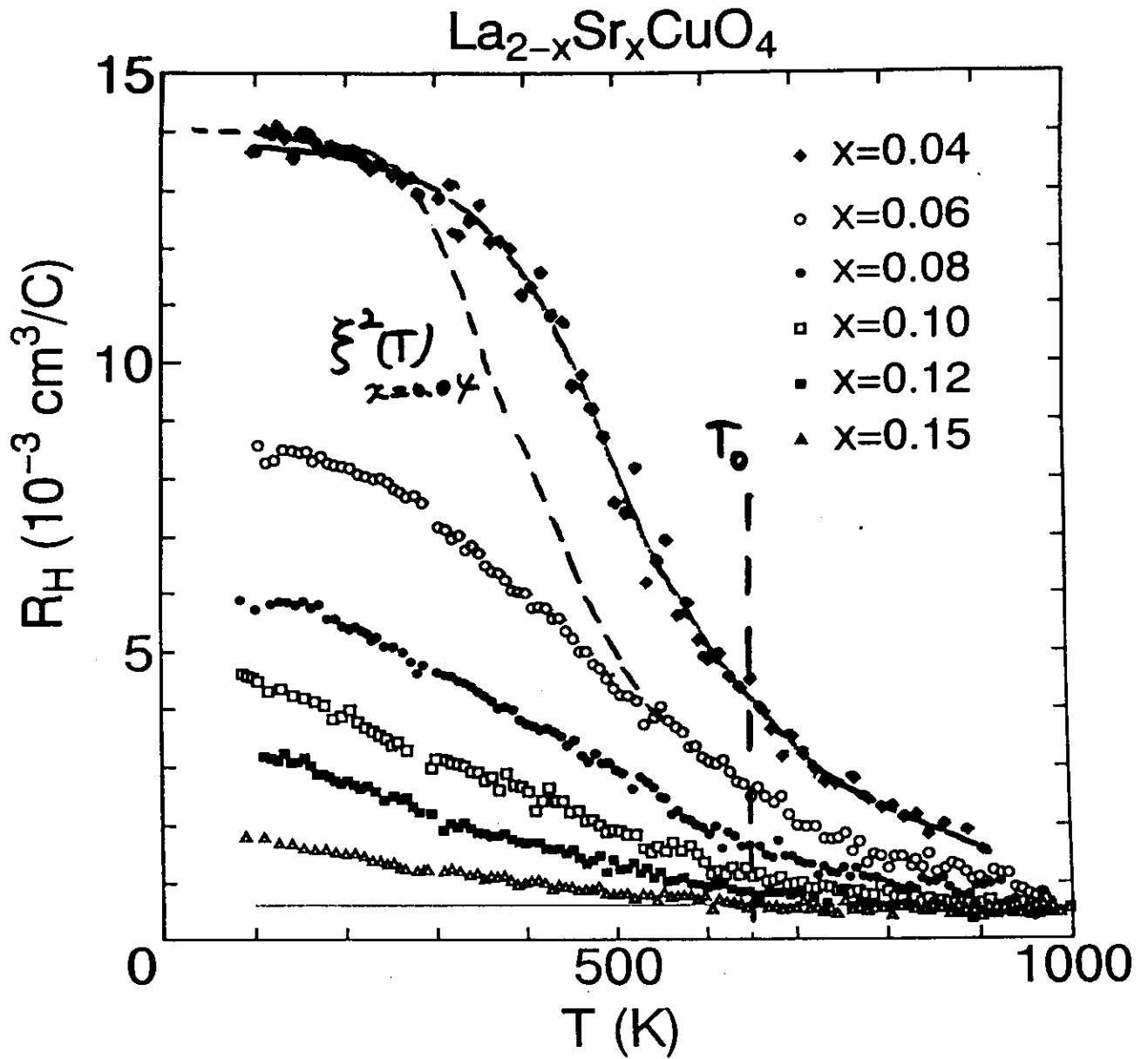
Loram et al,  
Nishikawa et al,  
(Nagoya)



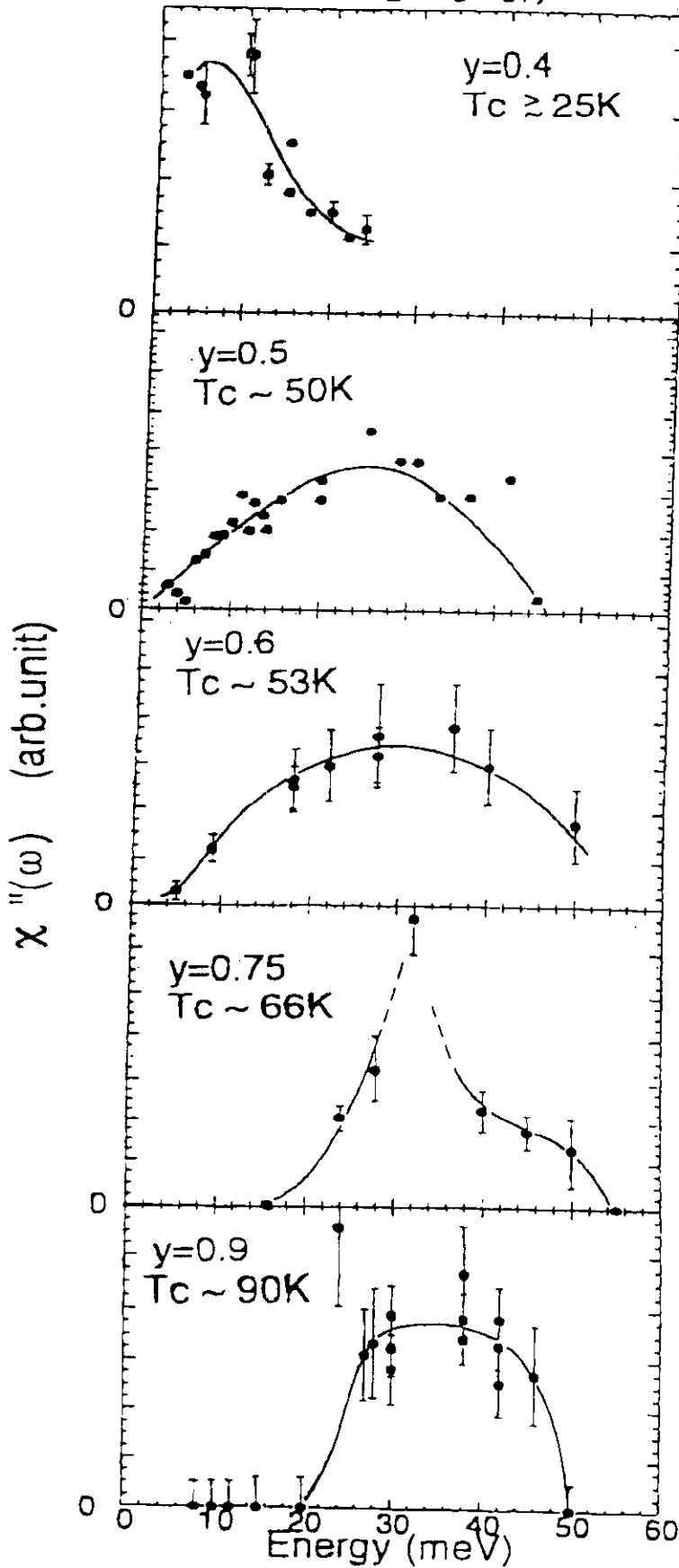
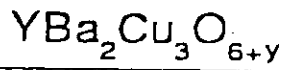
Loram et al.

$\gamma$  decreases as the system approaches  
M-I phase boundary.

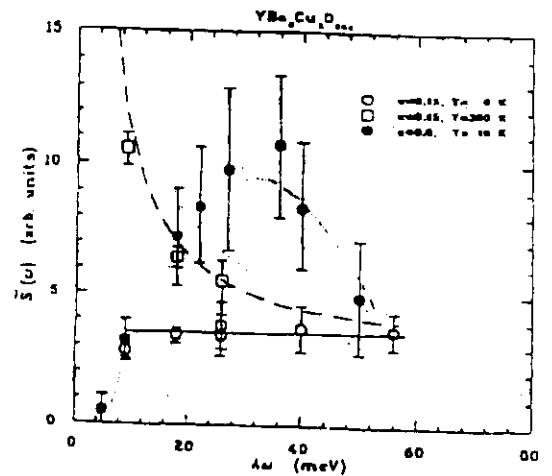
What is the origin of the anomalous phase ?



$\xi^2(T)$  : G. Shirane et al.



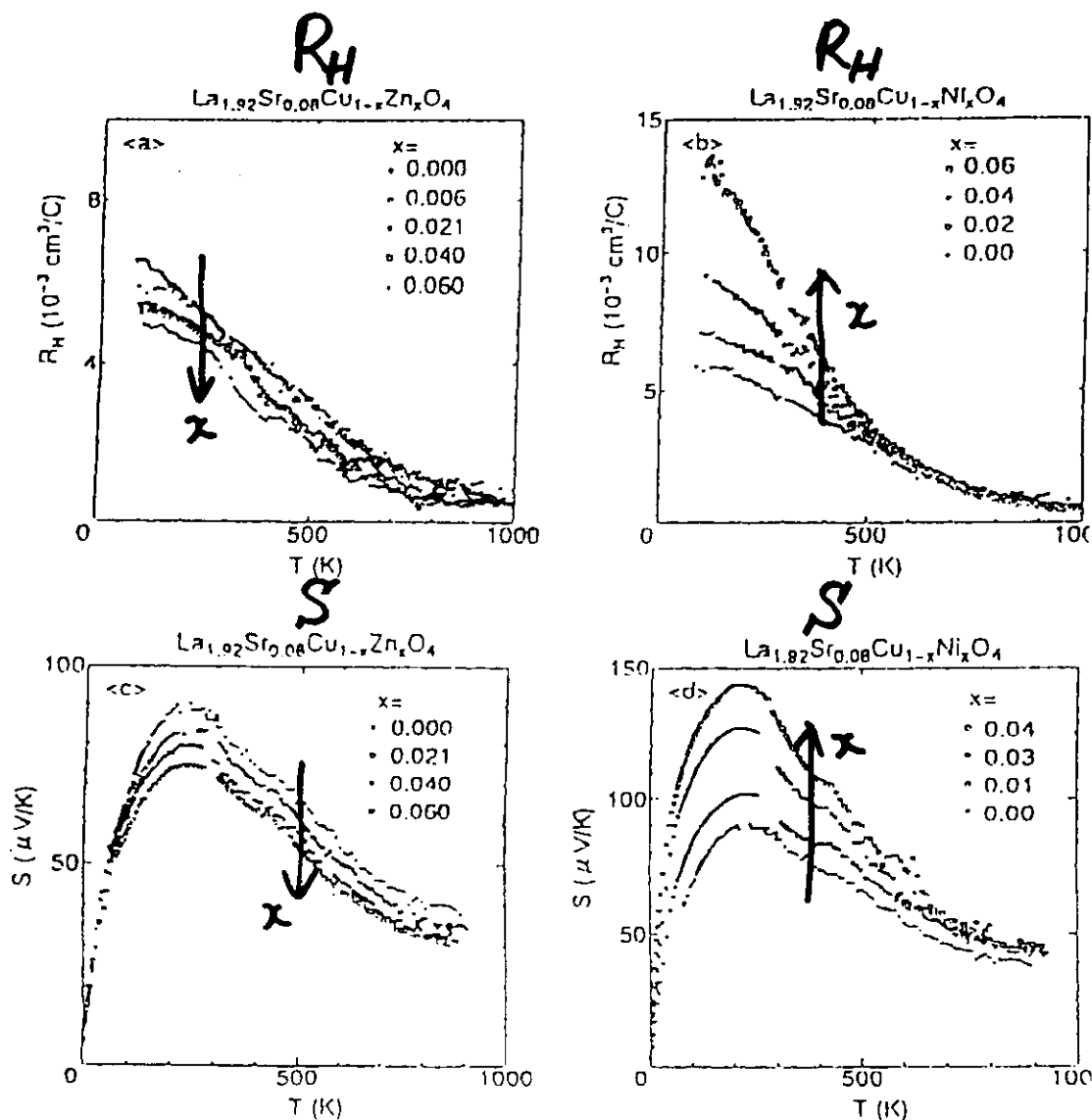
$\chi''(\omega)$  of YBCO  
our data



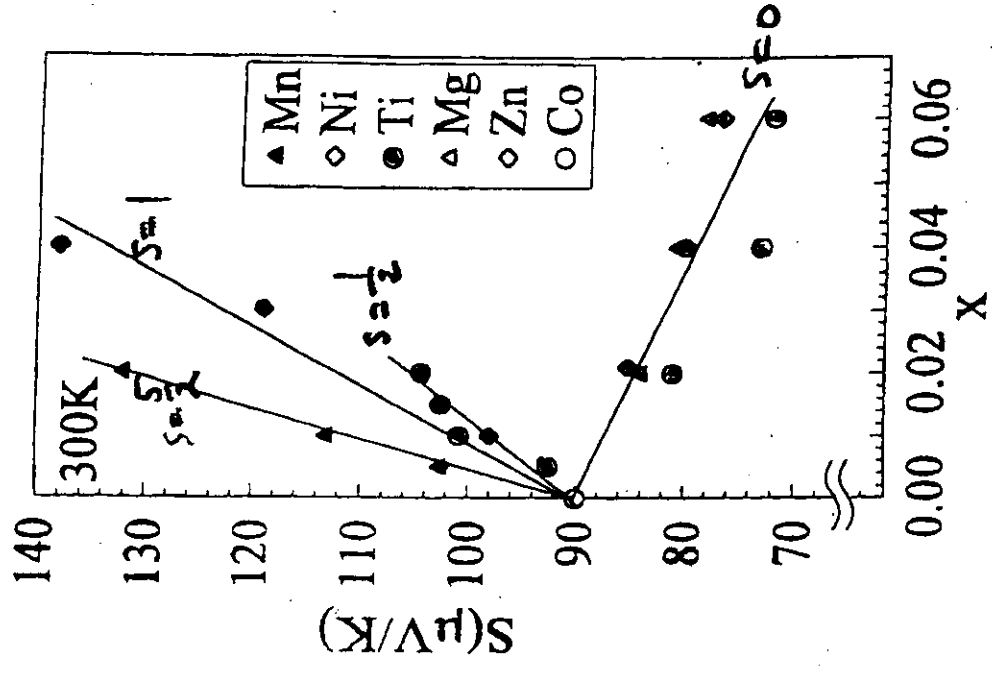
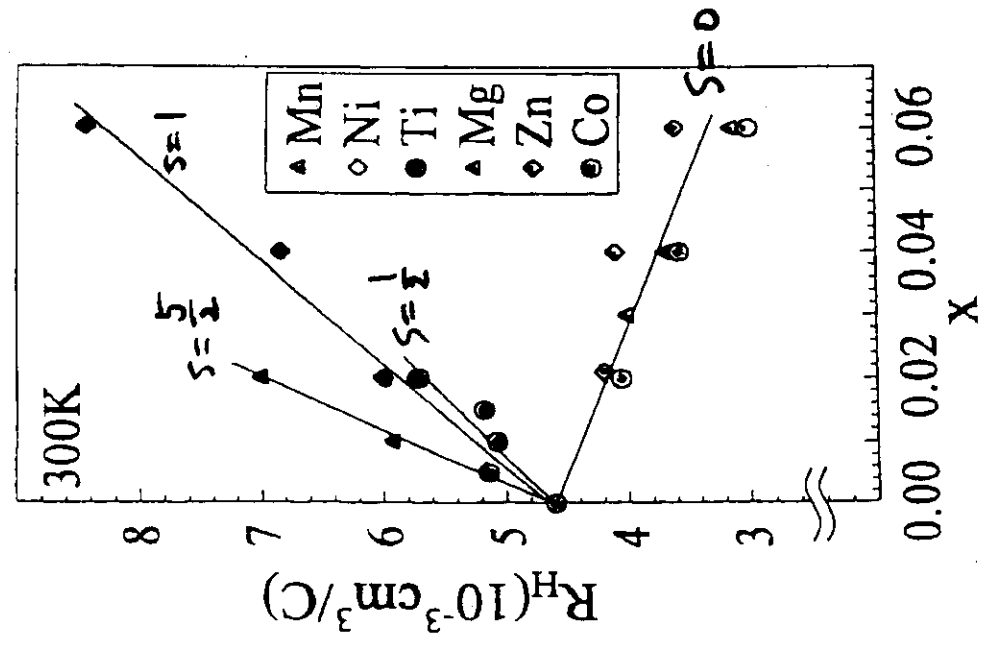
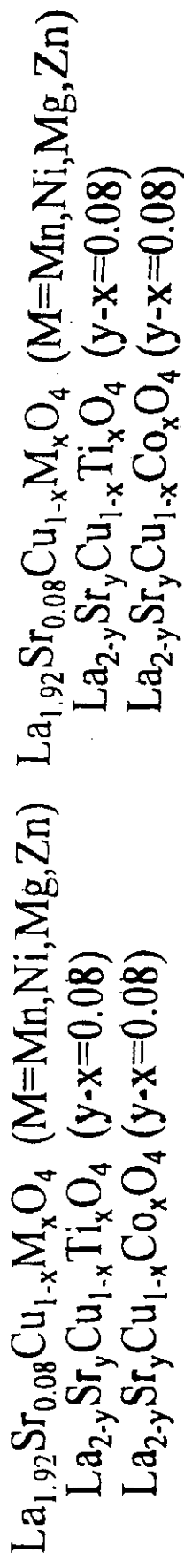
# Doping effect on the anomalous behavior of $S$ and $R_H$ (La 214)

Zn-doped ( $S=0$ )

Ni-doped ( $S=1$ )

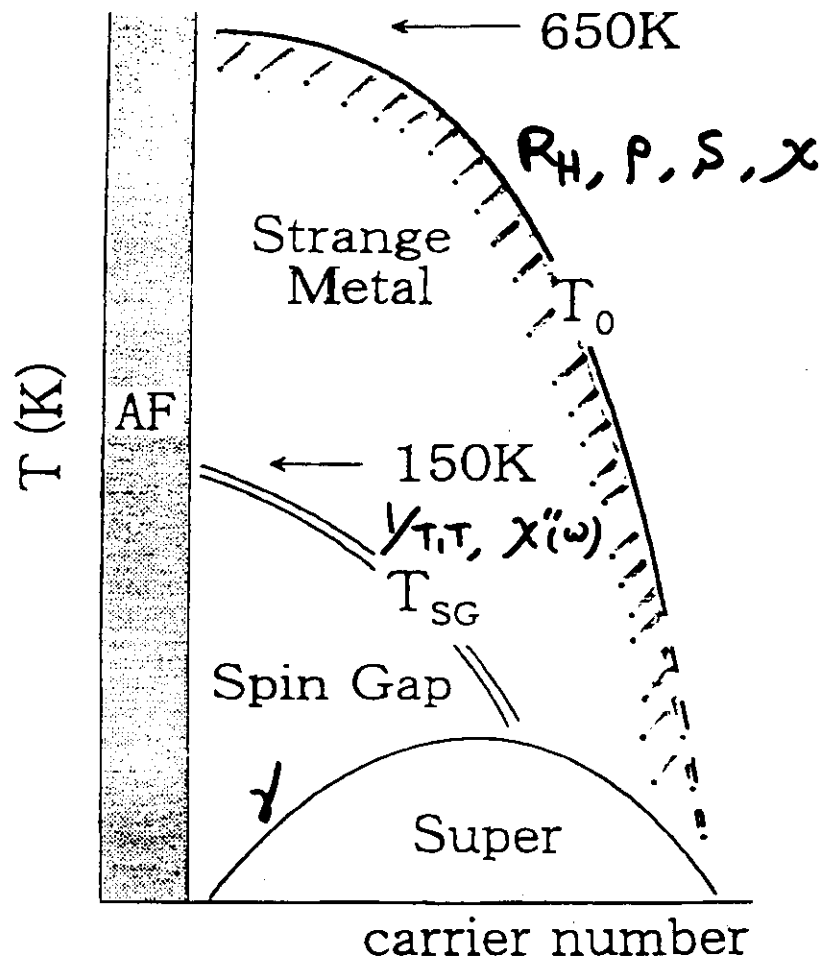


J. Takeda et al.  
(Nagoya)



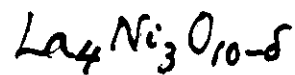
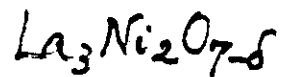
Enhancement of anomalous behavior of  $R_H$  and  $S$  by magnetic impurities  
 suppression of anomalous behavior of  $R_H$  and  $S$  by nonmagnetic impurities

# Anomalous metallic phase



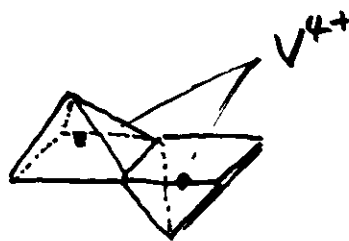
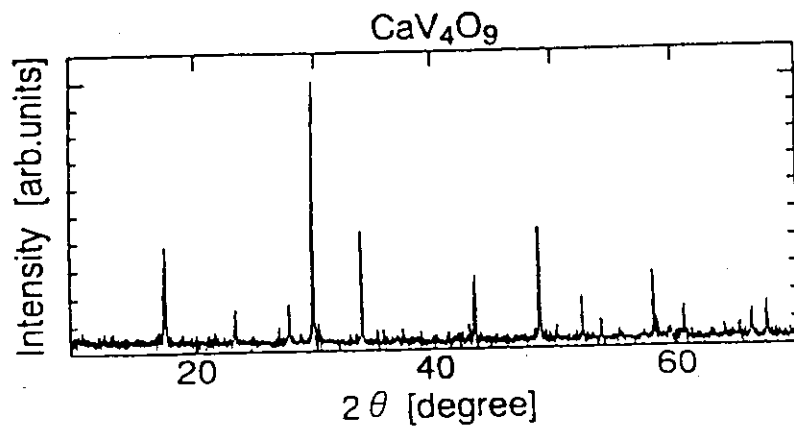
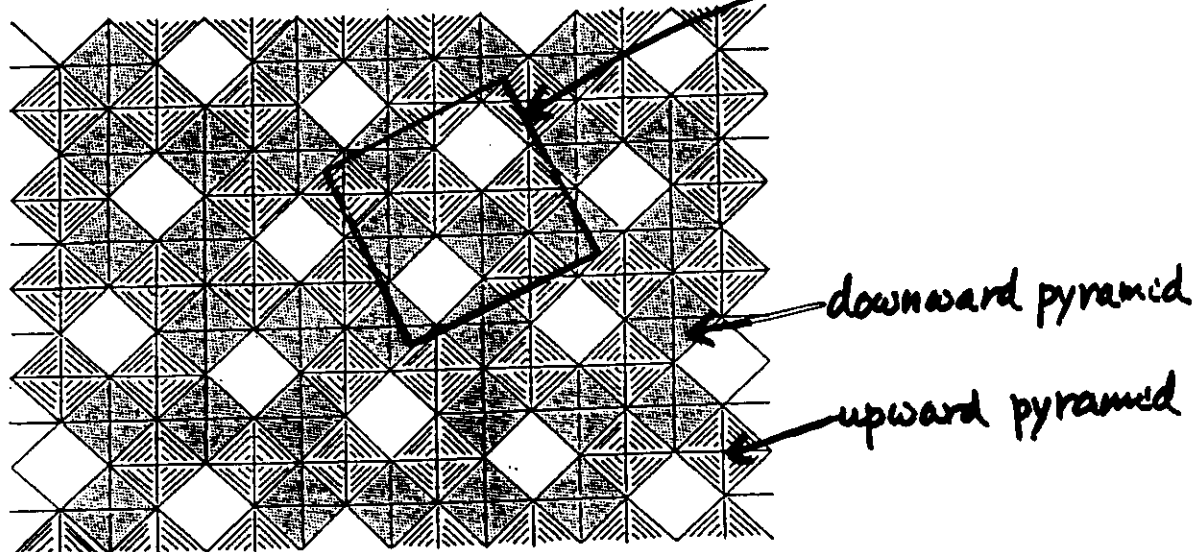
Characteristics of 2D electrons with  
 large spatial spin correlation,  
 low energy spin fluctuations,  
 & no long range magnetic ordering.  
 (↑ near the Mott insulating phase)

In order to clarify what characteristics of electron systems with 2D nature and near the Mott M-I phase boundary are relevant to High- $T_c$ , studies on non-Cu system with similar characteristics are interesting.

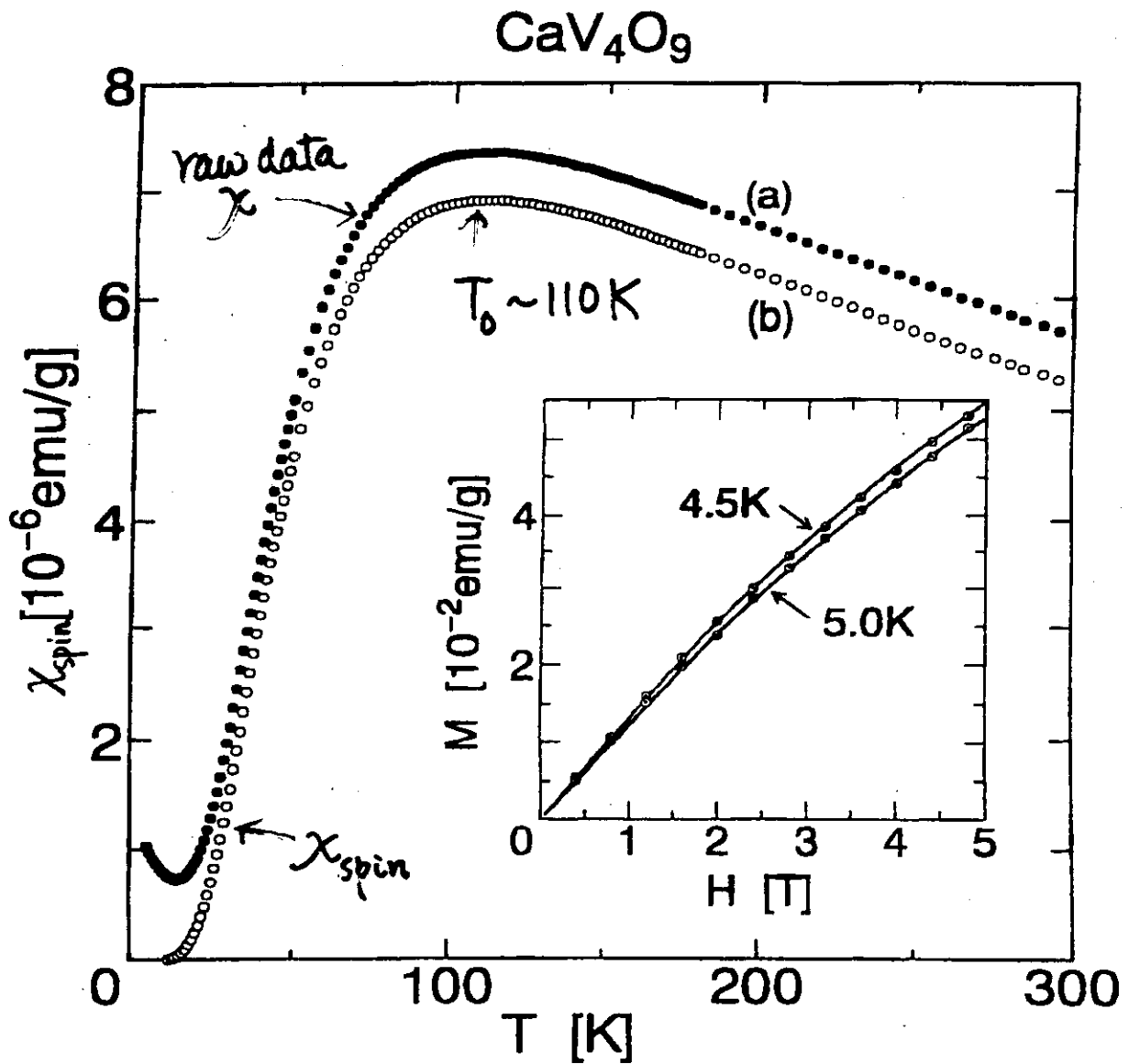




$\text{V}^{4+}$ ,  $S=1/2$ , 2D layered unit cell



# static magnetic susceptibility

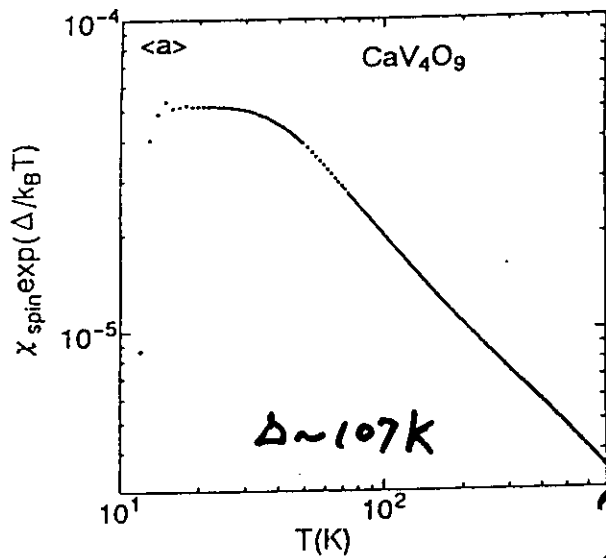


$$\chi = \chi_{spin} + \chi_0 + \alpha B_J \left( \frac{\mu H}{k_B T} \right)$$

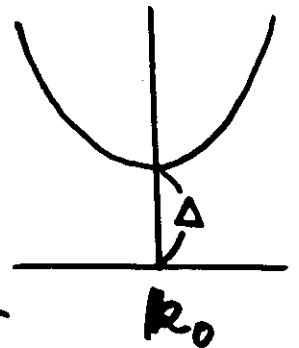
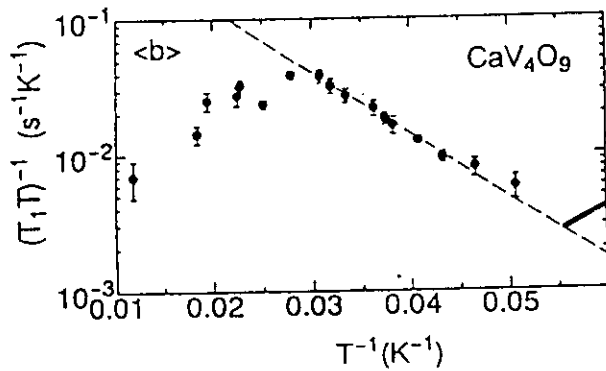
↑  
J=1

Taniguchi et al.

$\chi_{spin} e^{\Delta}$



$1/(T_i T)$



$$\chi_{spin} \sim \text{const.} \times e^{-\Delta/k_B T}$$

$$1/(T_i T) \sim \text{const.} \times e^{-\Delta/k_B T}$$



2D spin gap system!

# T dep. of $\chi$ and $1/T_1$

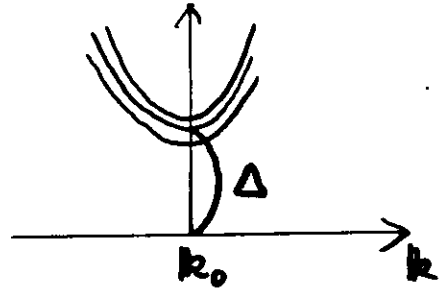
(i)  $\chi \sim T^{d/2-1} e^{-\beta\Delta}$  ( $d$ : dim) ( $T \ll \Delta$ )

$$Z \cong \prod_k \left\{ 1 + e^{-\beta(\epsilon_k + g\mu_B H)} + e^{-\beta\epsilon_k} + e^{-\beta(\epsilon_k - g\mu_B H)} \right\}$$

$$F = -\frac{1}{\beta} \ln Z \sim -\frac{1}{\beta} \sum_k \left\{ 1 + 2 \cosh(\beta g\mu_B H) \right\} e^{-\beta\epsilon_k}$$

$$\sum_k \rightarrow \int N(\epsilon) d\epsilon$$

$N(\epsilon)$ :  $d$ -dependent



(ii)  $\frac{1}{T_1 T} \sim e^{-\beta\Delta}$  ( $d=2$ )

excited state  $|k\rangle$

nuclear spin relaxation ← scattering

$$|k\rangle \rightarrow |k+q\rangle \quad q \sim 0$$

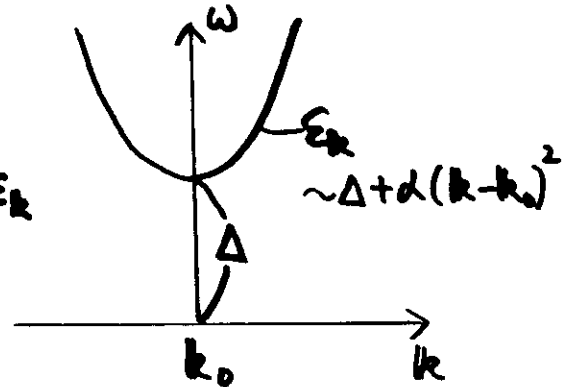
$$\frac{1}{T_1} \propto \sum_{q \neq 0} S(q, \omega)$$

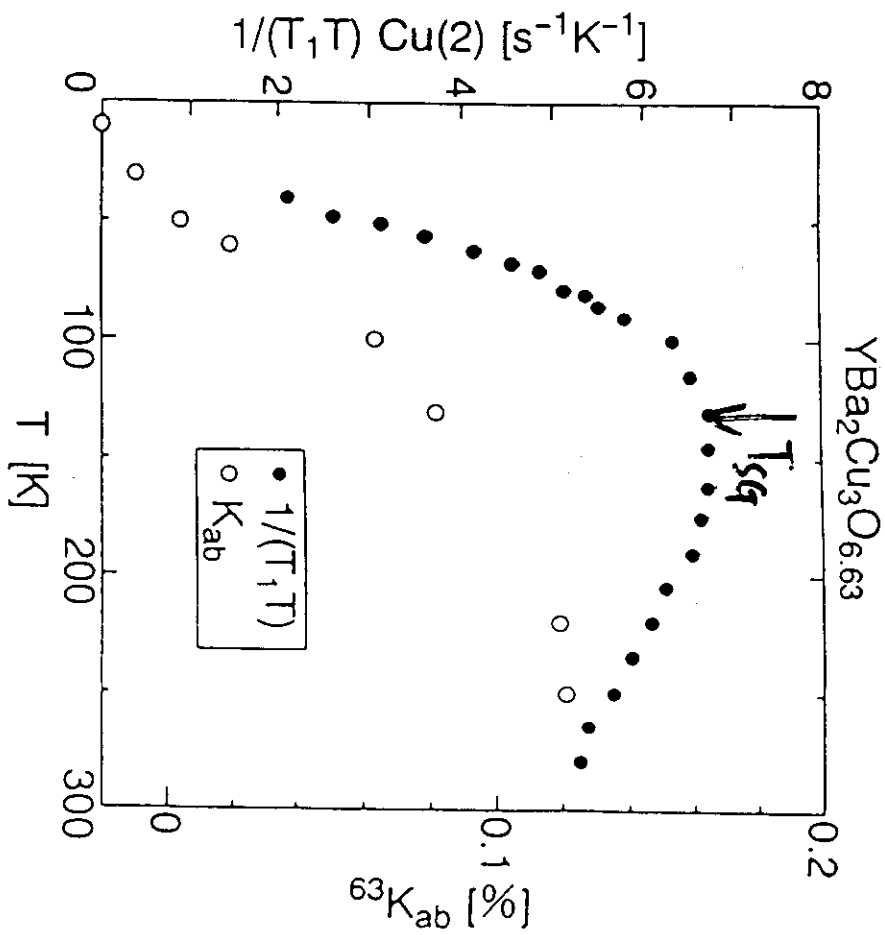
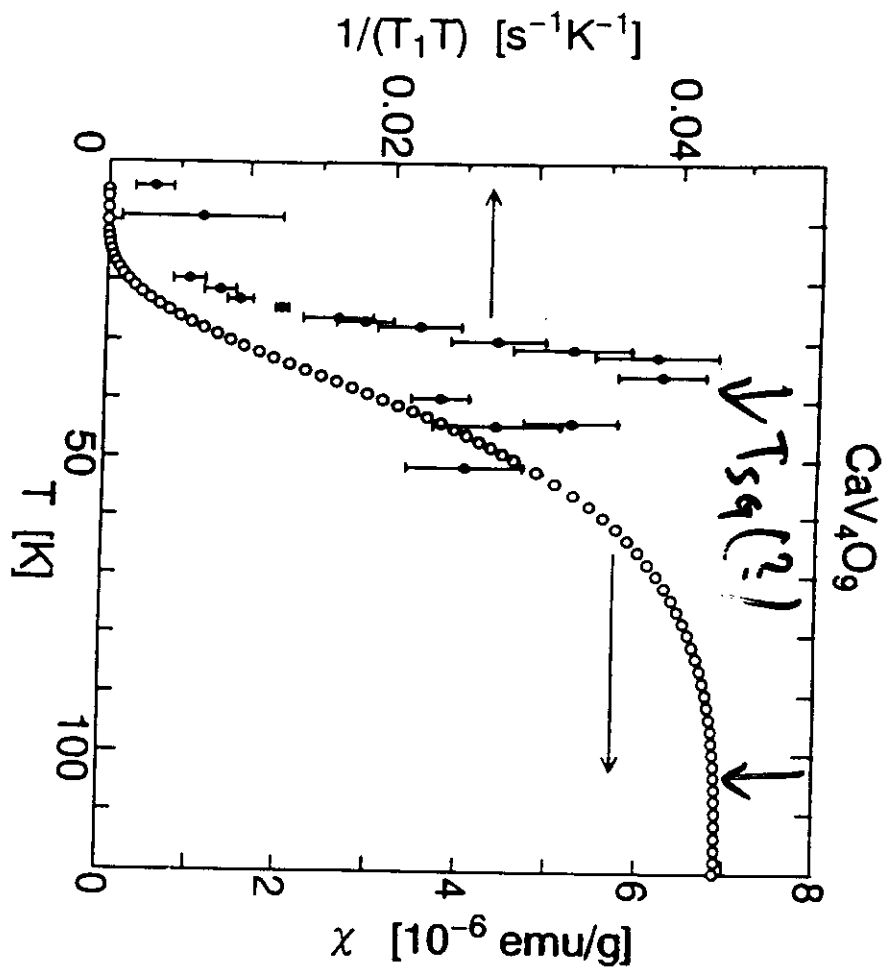
$$\sim \sum_{q, k} |\langle k+q | S_q | k \rangle|^2 \times \delta(\epsilon_{k+q} - \epsilon_k - \hbar\omega) e^{-\beta\epsilon_k}$$

$$\omega_0 \sim 0$$

$$\sim \text{const.} \times \sum_k e^{-\beta\epsilon_k}$$

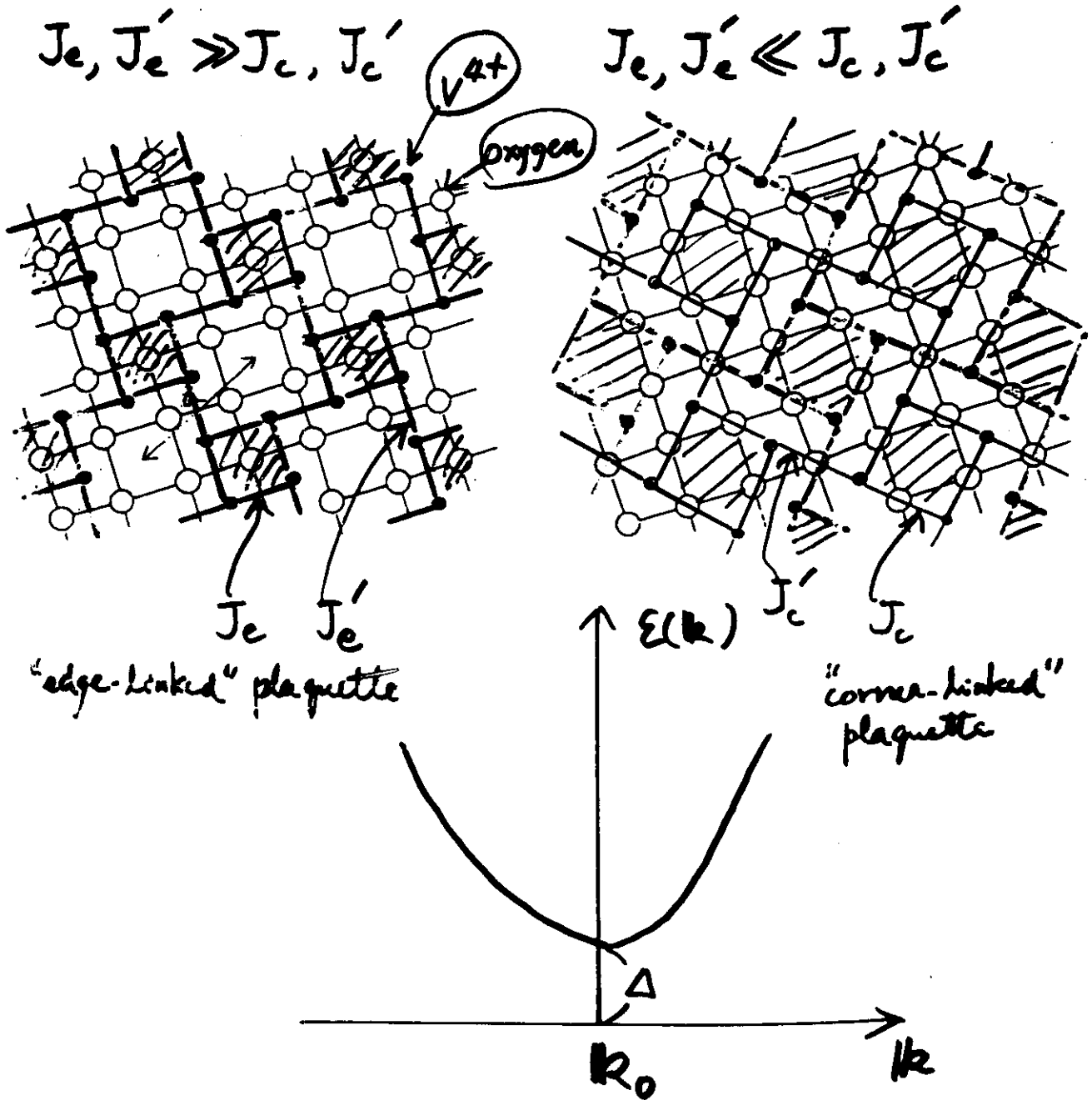
$$\sim \int_{\Delta}^{\infty} e^{-\beta\epsilon} d\epsilon \sim \frac{1}{\beta} e^{-\beta\Delta}$$





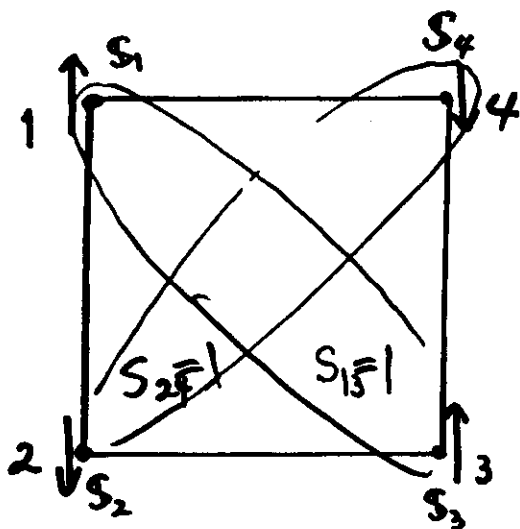
What is the origin of spin gap?

→ plaquette unit?



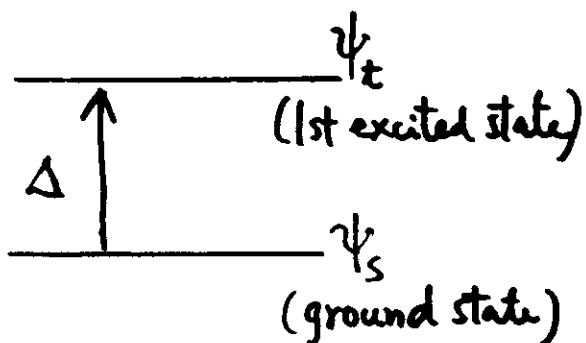
- where is  $k_0$  ?
- structure factor distribution for neutron magnetic scattering ?

# Isolated plaquette



$$S_{13} = S_1 + S_3$$

$$S_{24} = S_2 + S_4$$



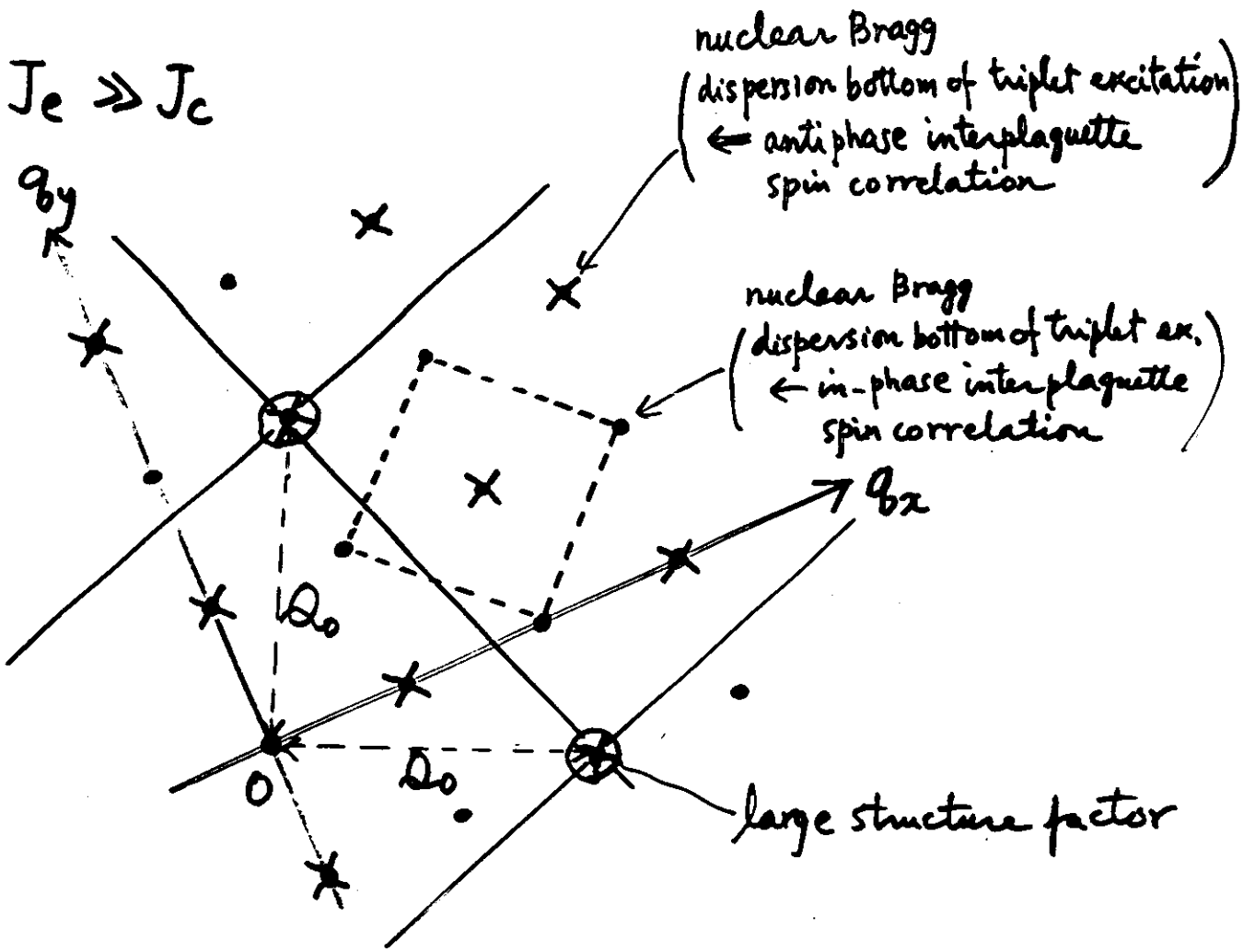
$$S=1 \quad (\leftarrow S_{13} + S_{24})$$

$$S=0 \quad (\leftarrow S_{13} \mp S_{24})$$

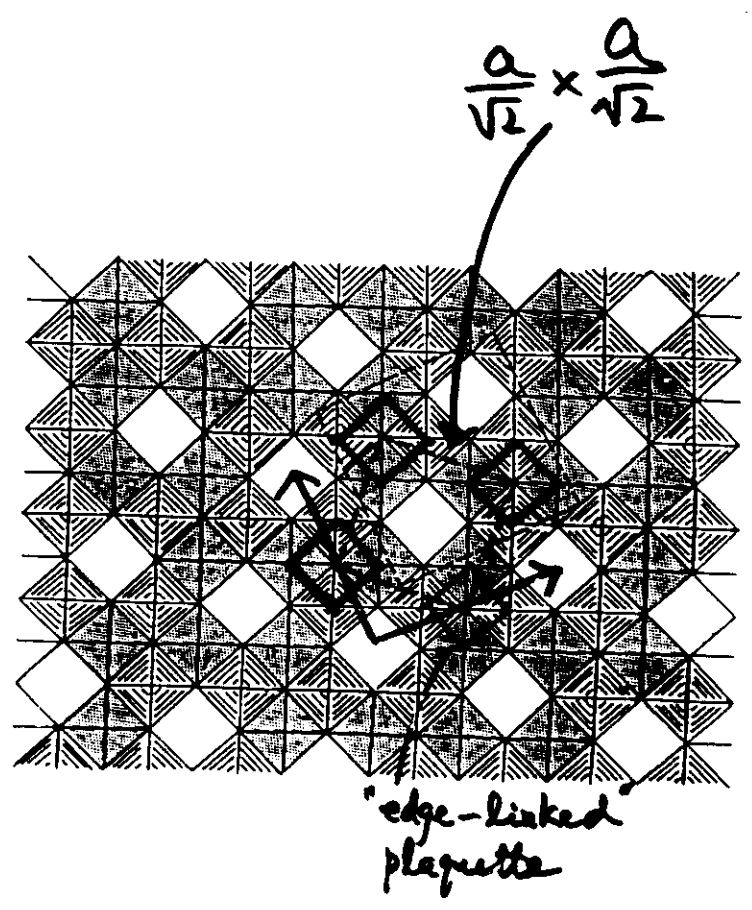
$$\psi_S \propto \left\{ \alpha_1 \beta_2 \alpha_3 \beta_4 + \beta_1 \alpha_2 \beta_3 \alpha_4 - \frac{1}{2} (\alpha_1 \alpha_2 \beta_3 \beta_4 + \beta_1 \alpha_2 \alpha_3 \beta_4 + \alpha_1 \beta_2 \beta_3 \alpha_4 + \beta_1 \beta_2 \alpha_3 \alpha_4) \right\}$$

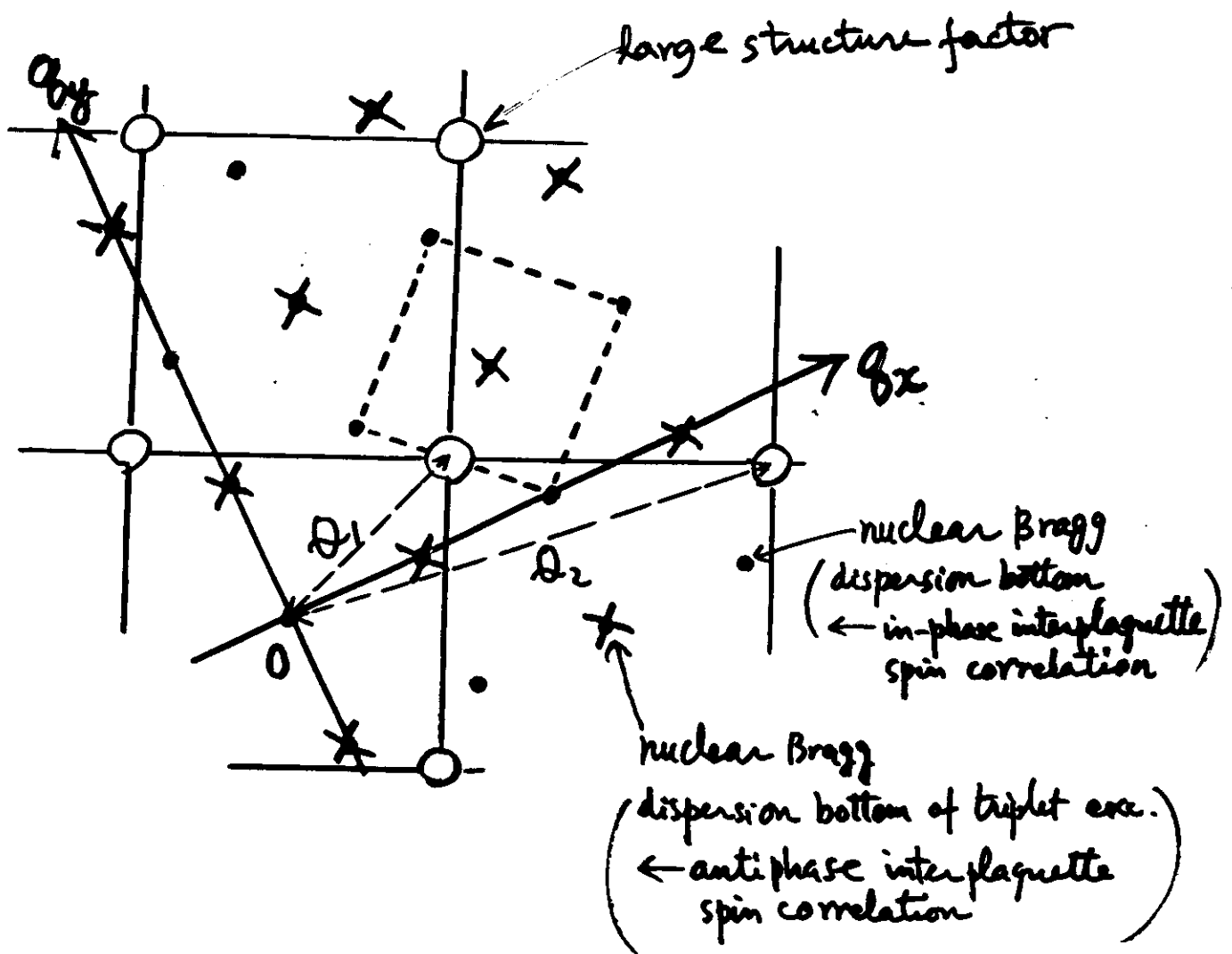
$$\psi_t \propto (\alpha_1 \beta_2 \beta_3 \beta_4 + \beta_1 \beta_2 \alpha_3 \beta_4 - \beta_1 \alpha_2 \beta_3 \beta_4 - \beta_1 \beta_2 \beta_3 \alpha_4)$$

$J_e \gg J_c$

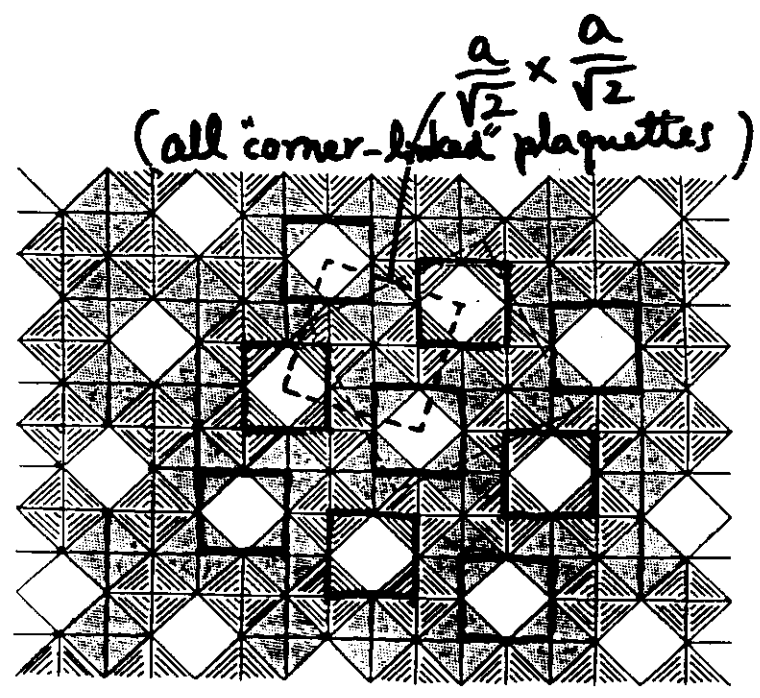


CASE I



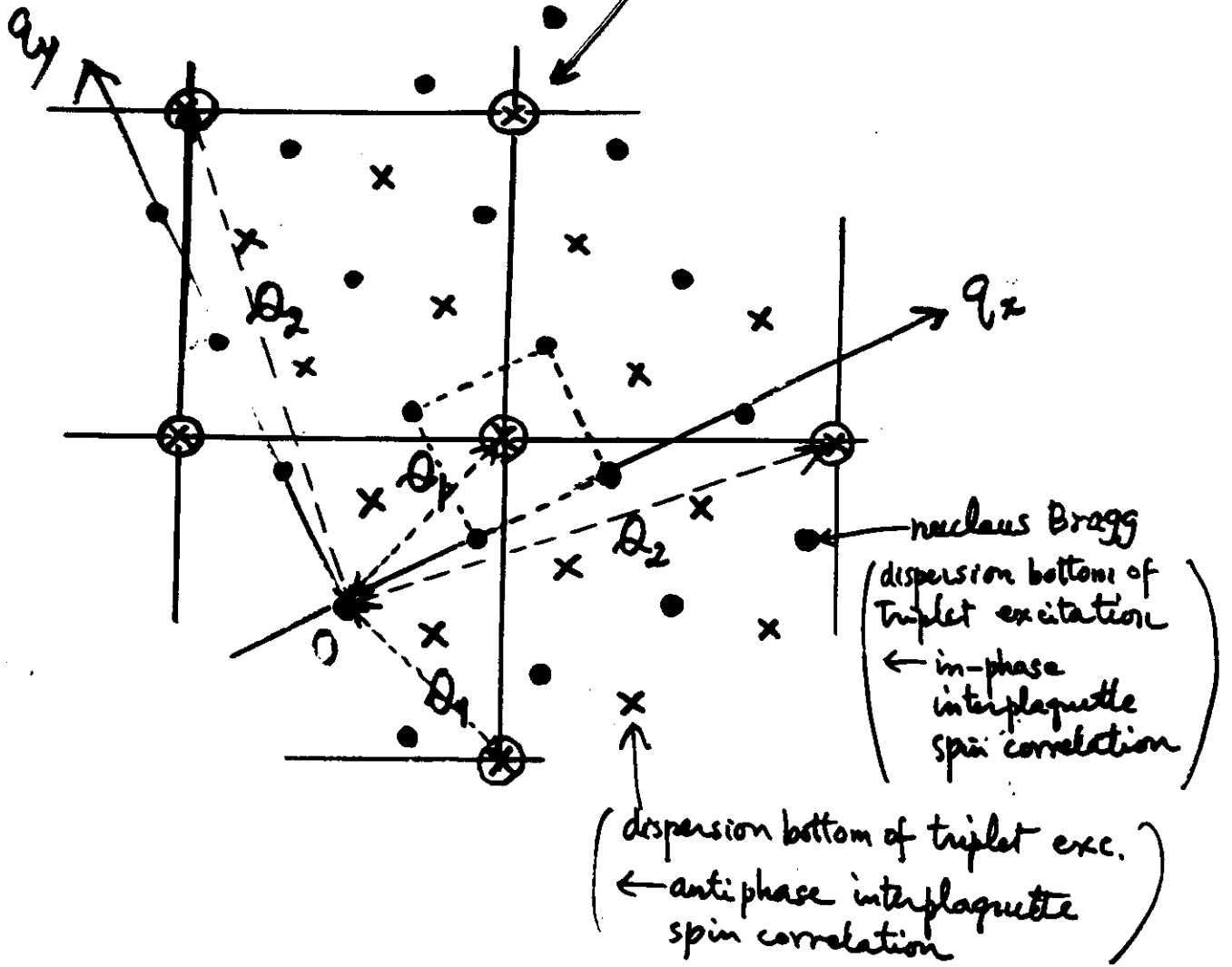


CASE III

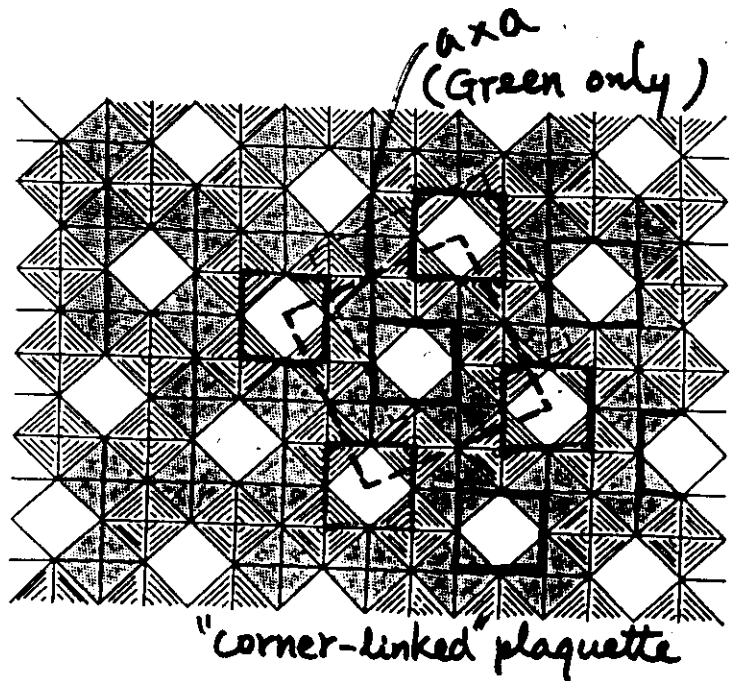


$J_c \gg J_e$

large structure factor



CASE II

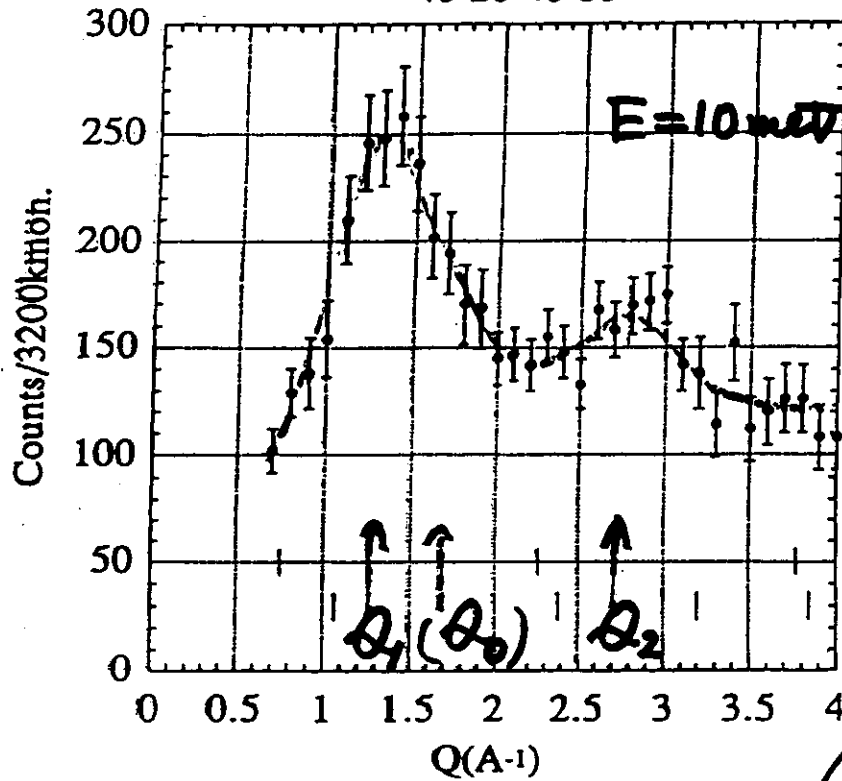


# Neutron Inelastic Measurements

powder

$E=10\text{meV}$  Q-scan 7K  $k_i=4.518\text{\AA}^{-1}$   
40-20-40-80

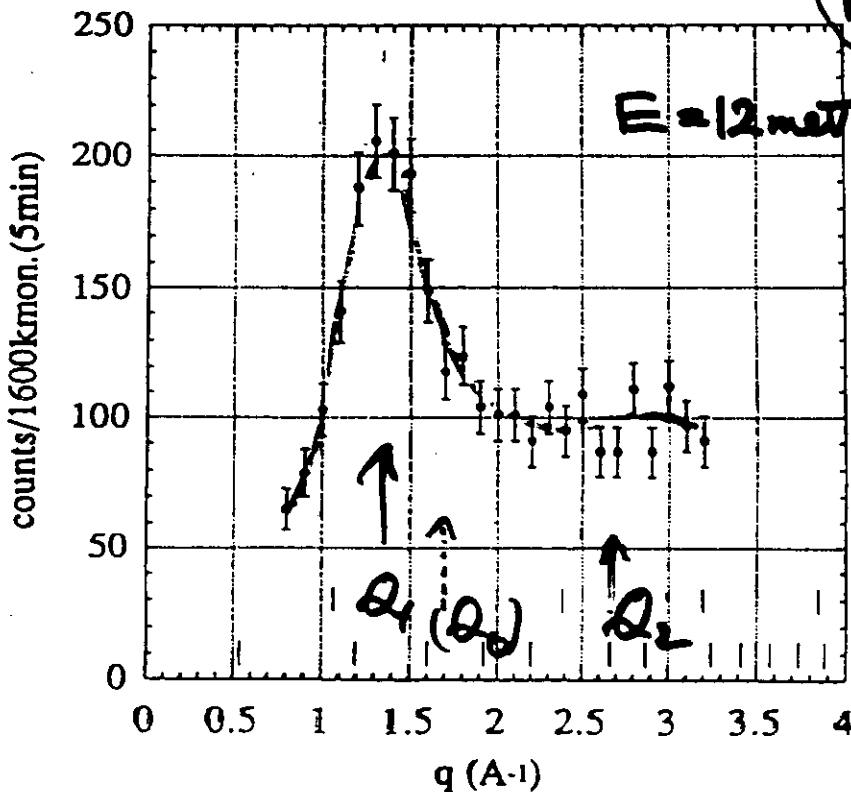
K. Kodama et al.



$E=12\text{meV}$  q-scan 7K  
40-pgf-20-s-40-80  $E_i=42.3\text{meV}$

corner-linked  
plaquette unit

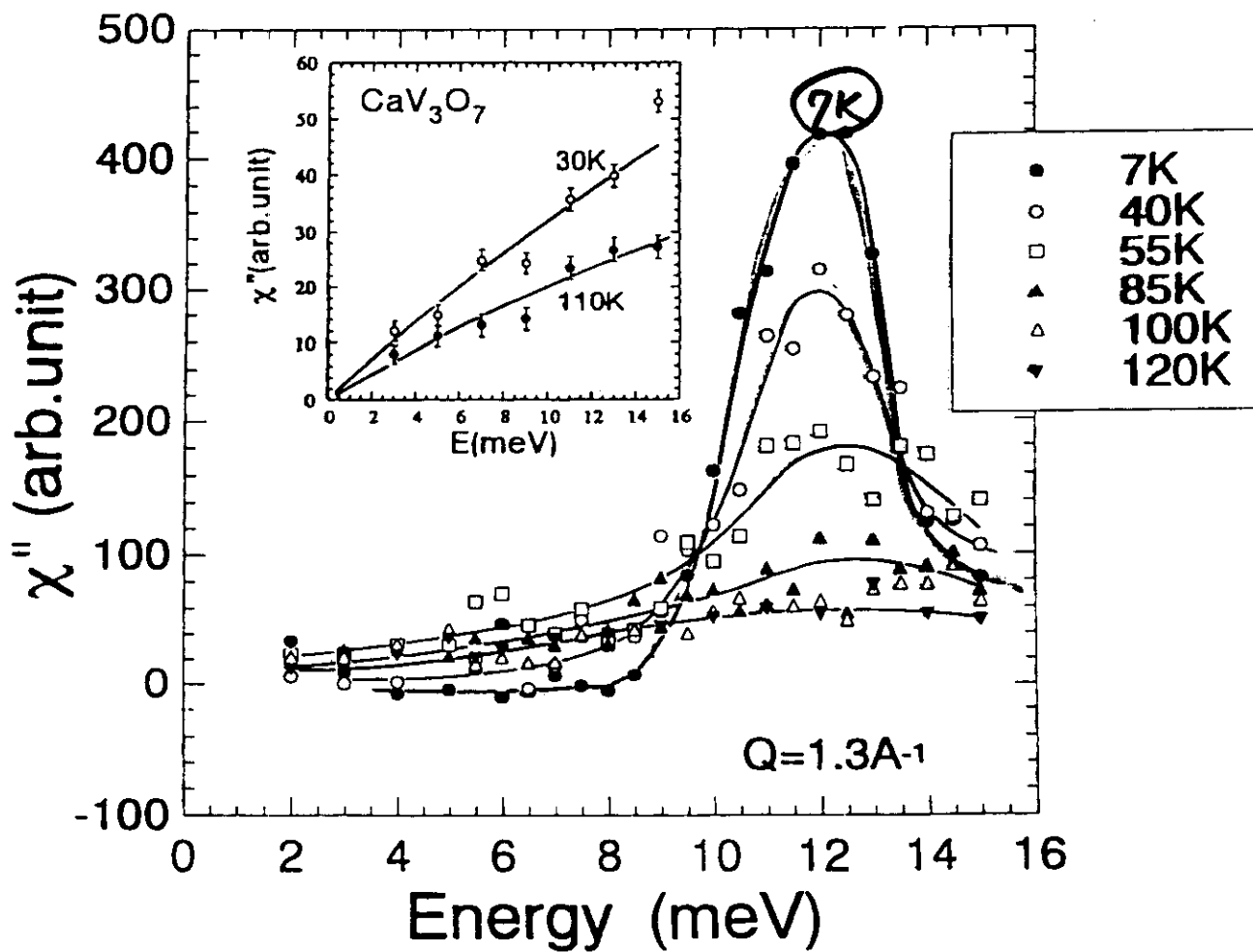
↓  
CASE II  
CASE III



powder data

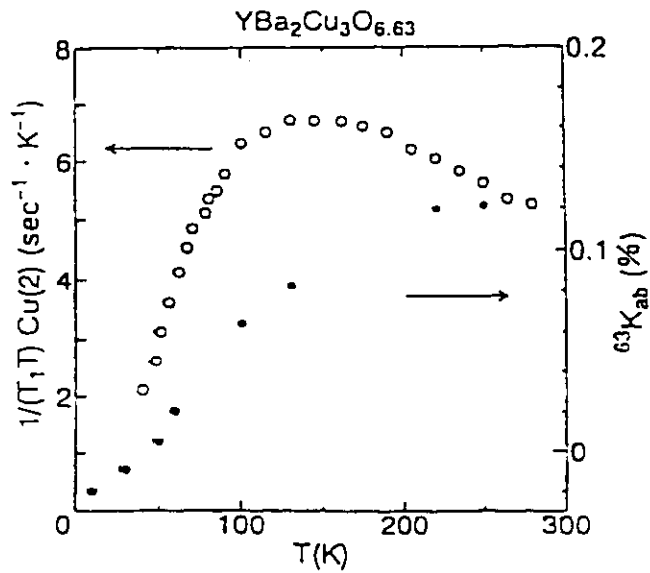
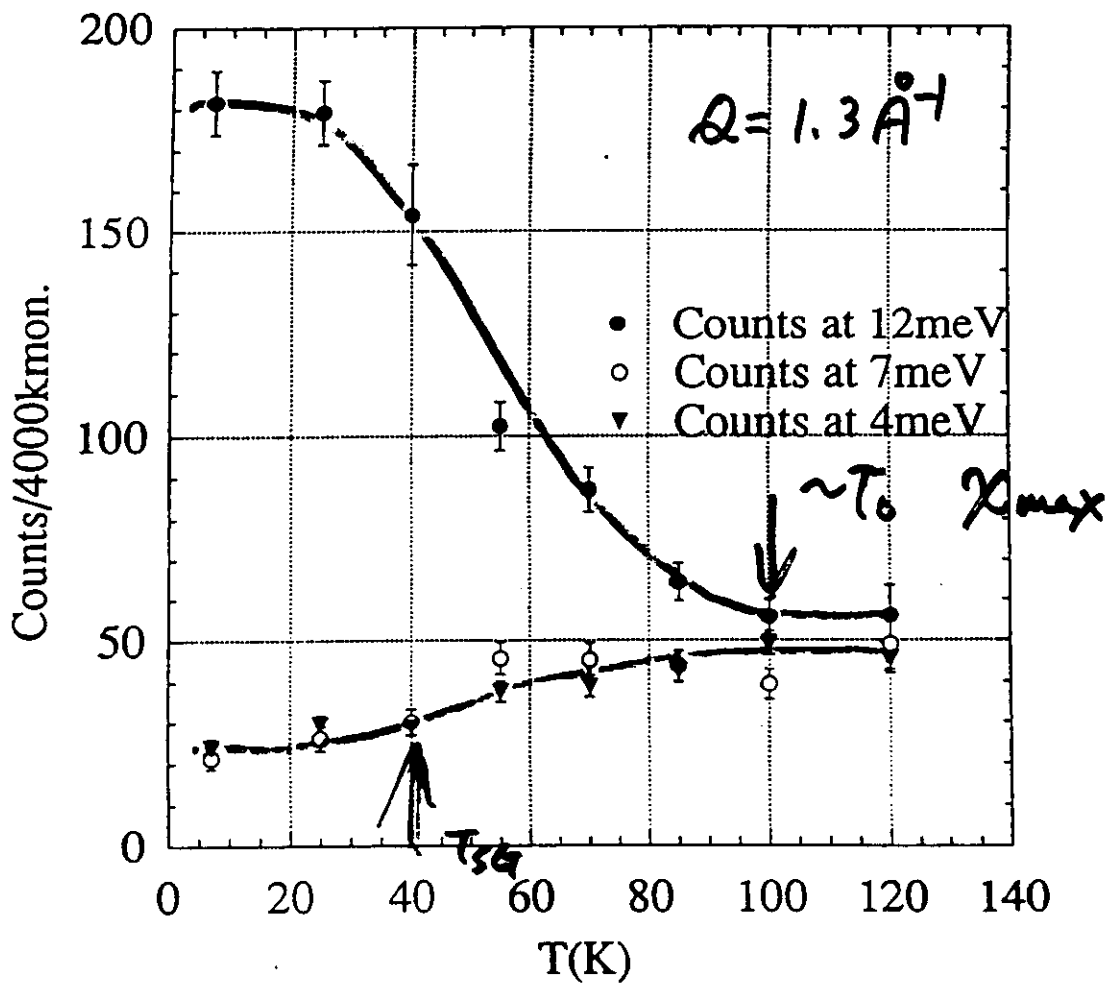
$$Q = 1.3 \text{ \AA}^{-1} (\sim Q_1)$$

### CaV<sub>4</sub>O<sub>9</sub>



K. Kodama et al.

T-dep at 12meV, 7meV, 4meV data



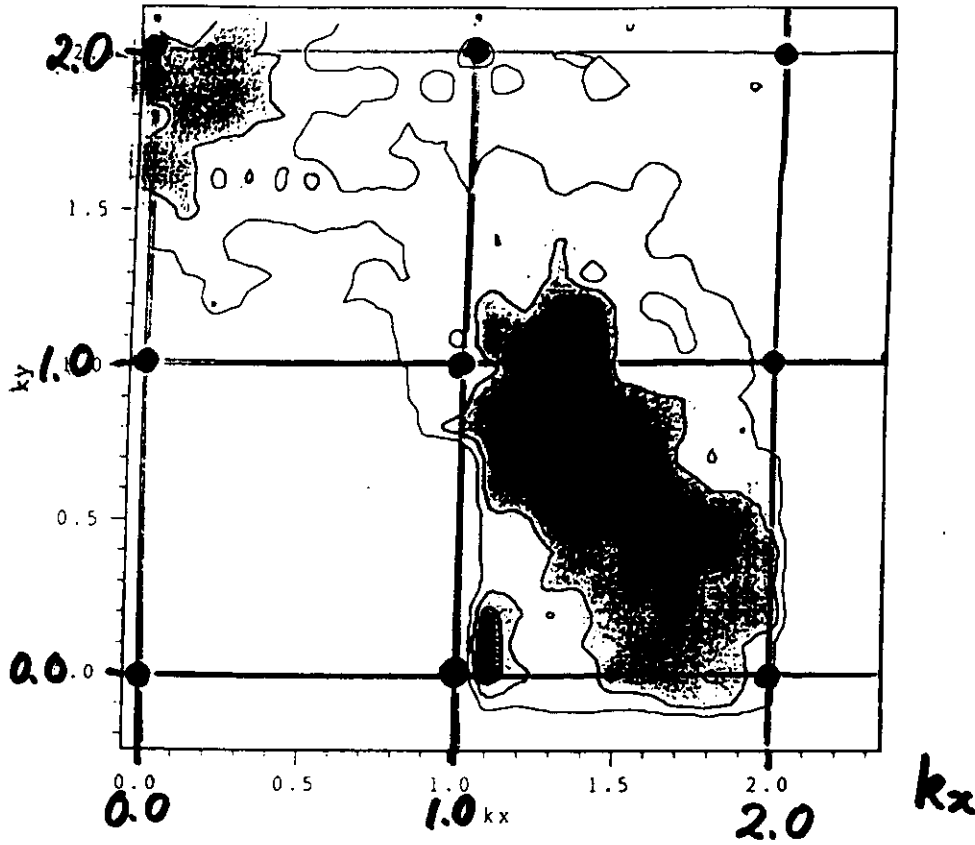
Single crystal study

$\text{CaV}_4\text{O}_9$

# Intensity map $k_y$

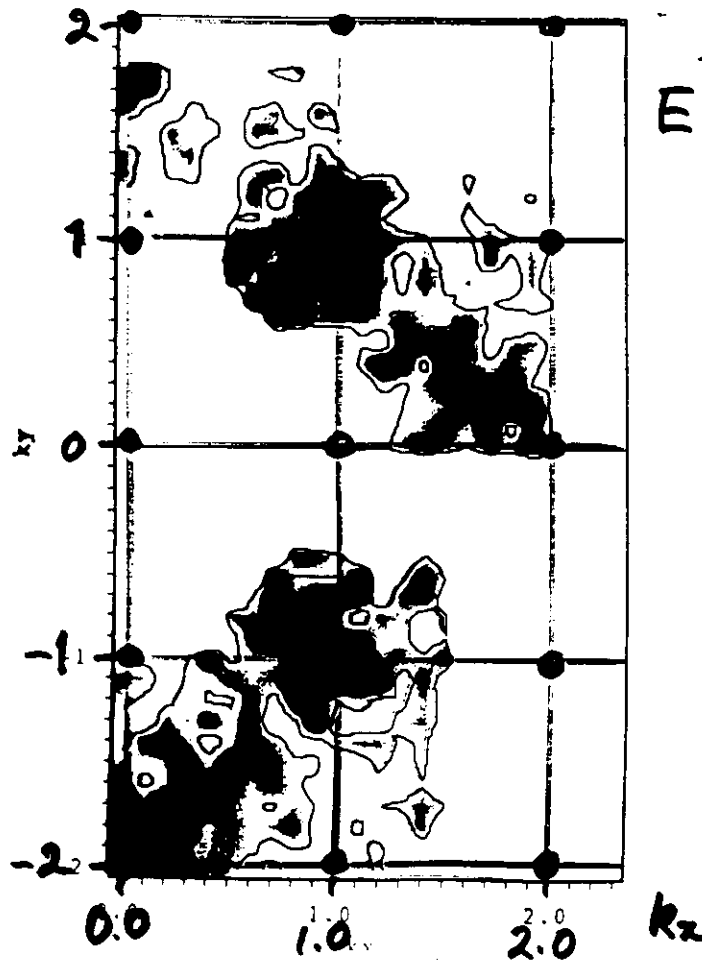
Q-scan 12meV-7K

$E = 12 \text{ meV}$



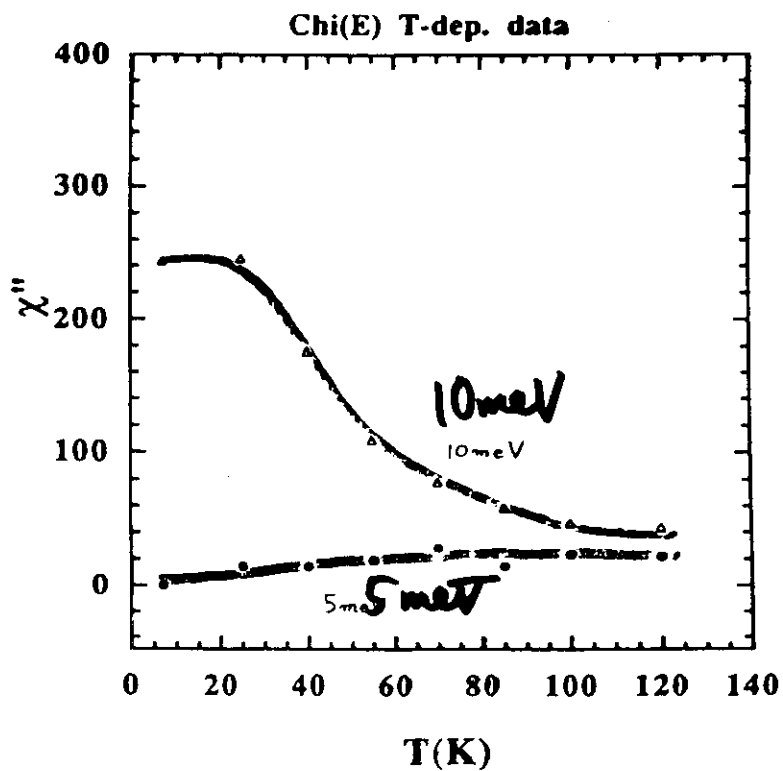
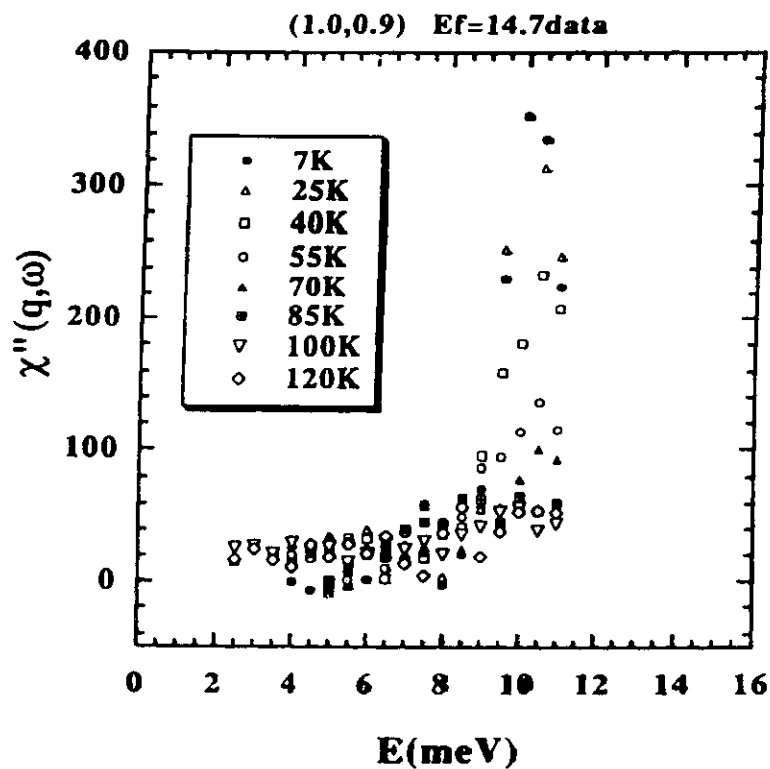
Q-scan 9meV-7K

$E = 9 \text{ meV}$



K. Kodama et al.

# Single crystal data

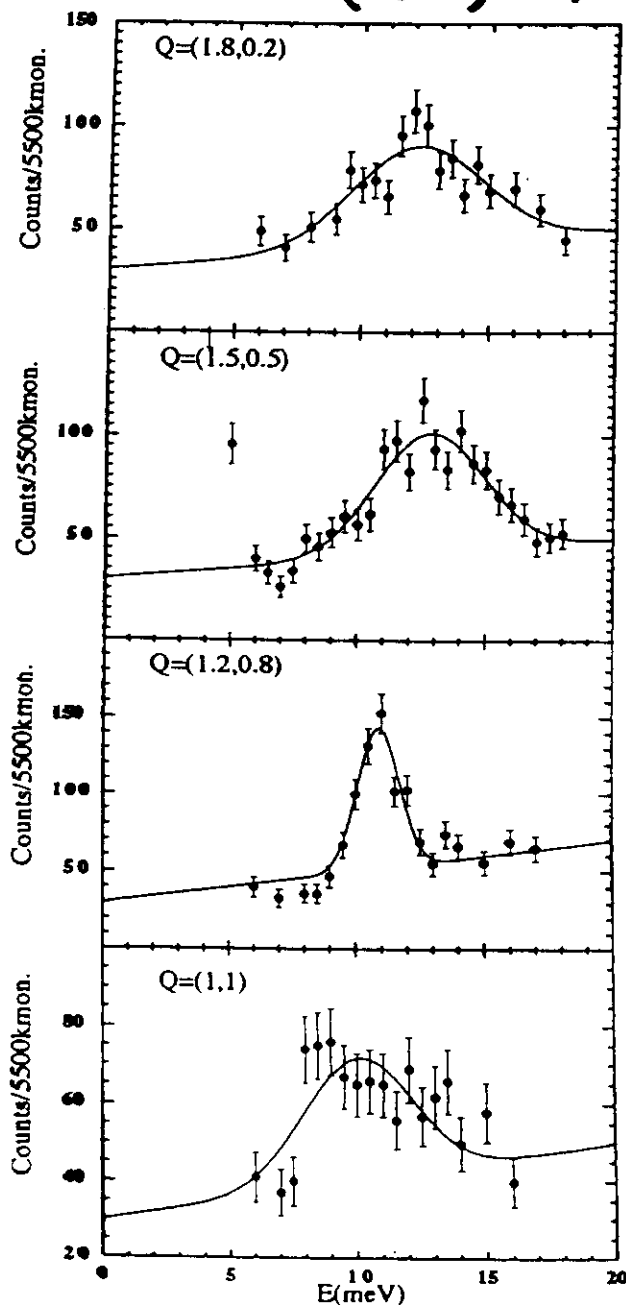
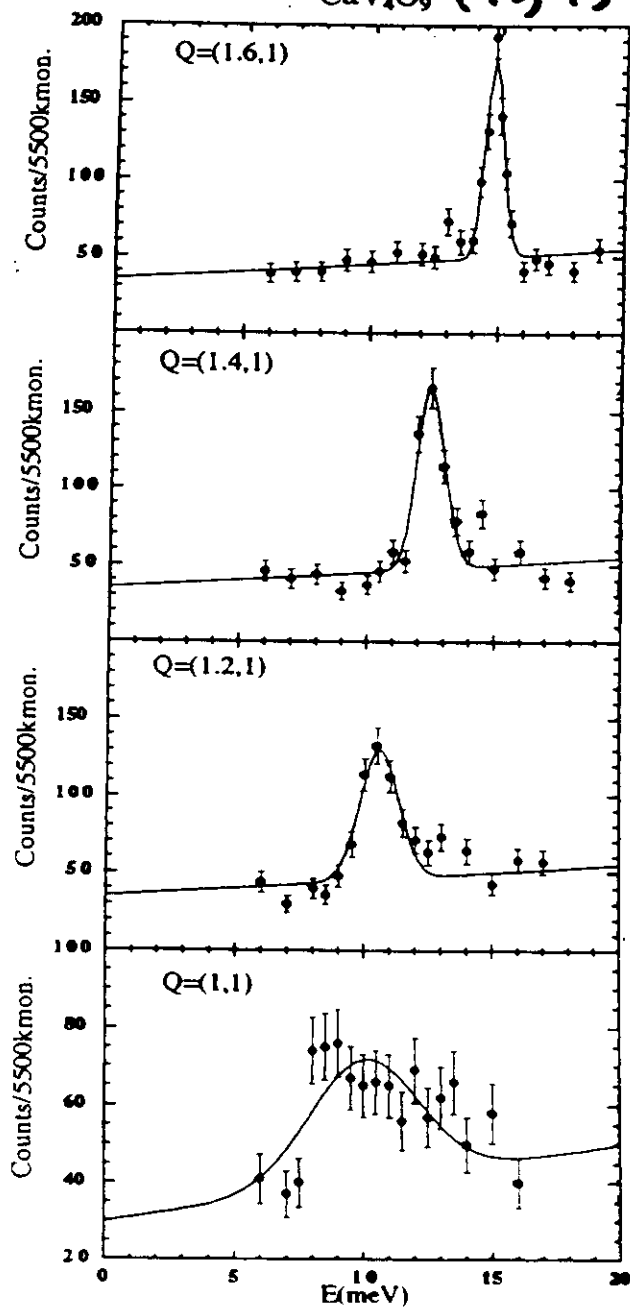


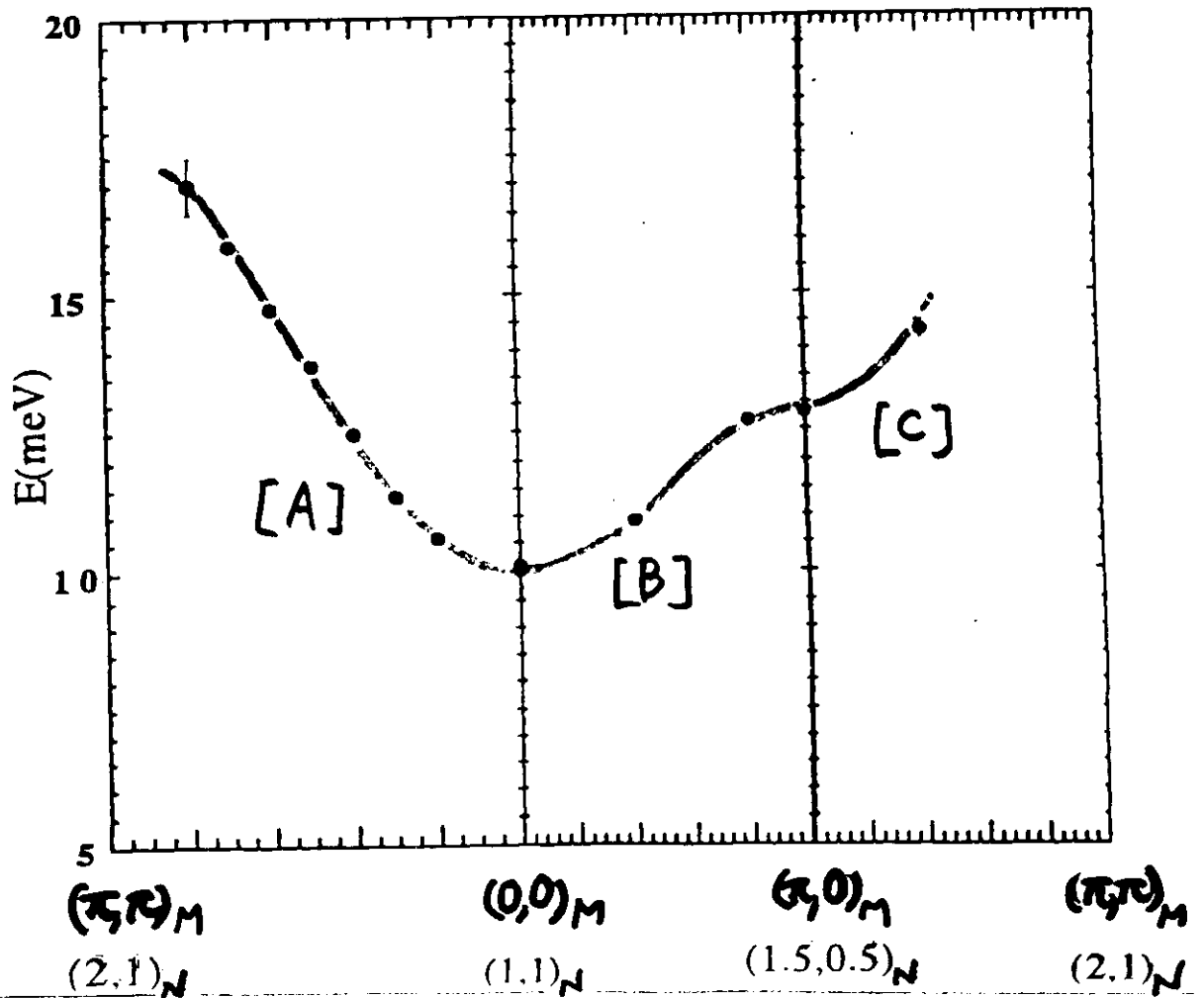
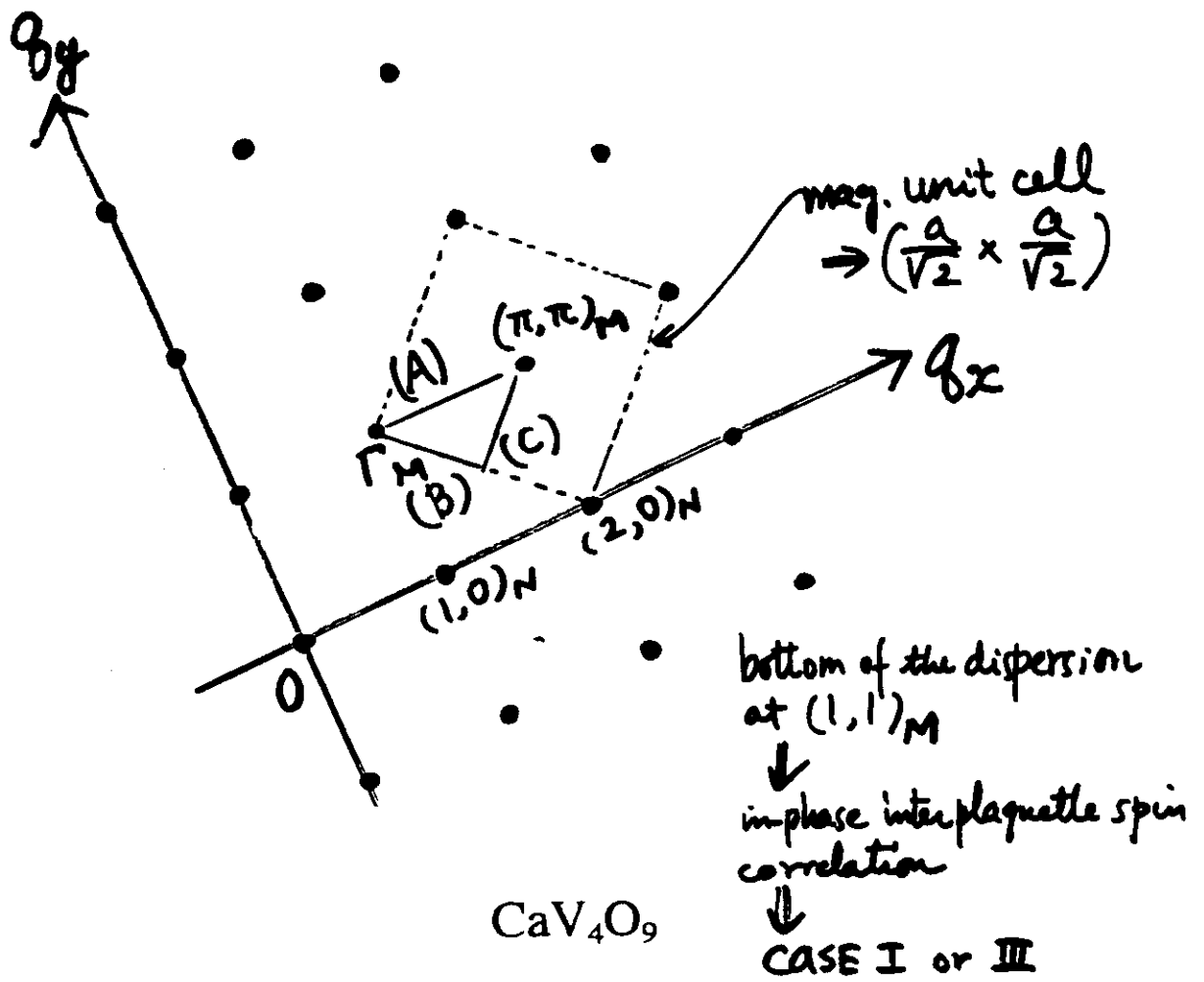
essentially similar to corresponding powder data

# Example results of dispersion study

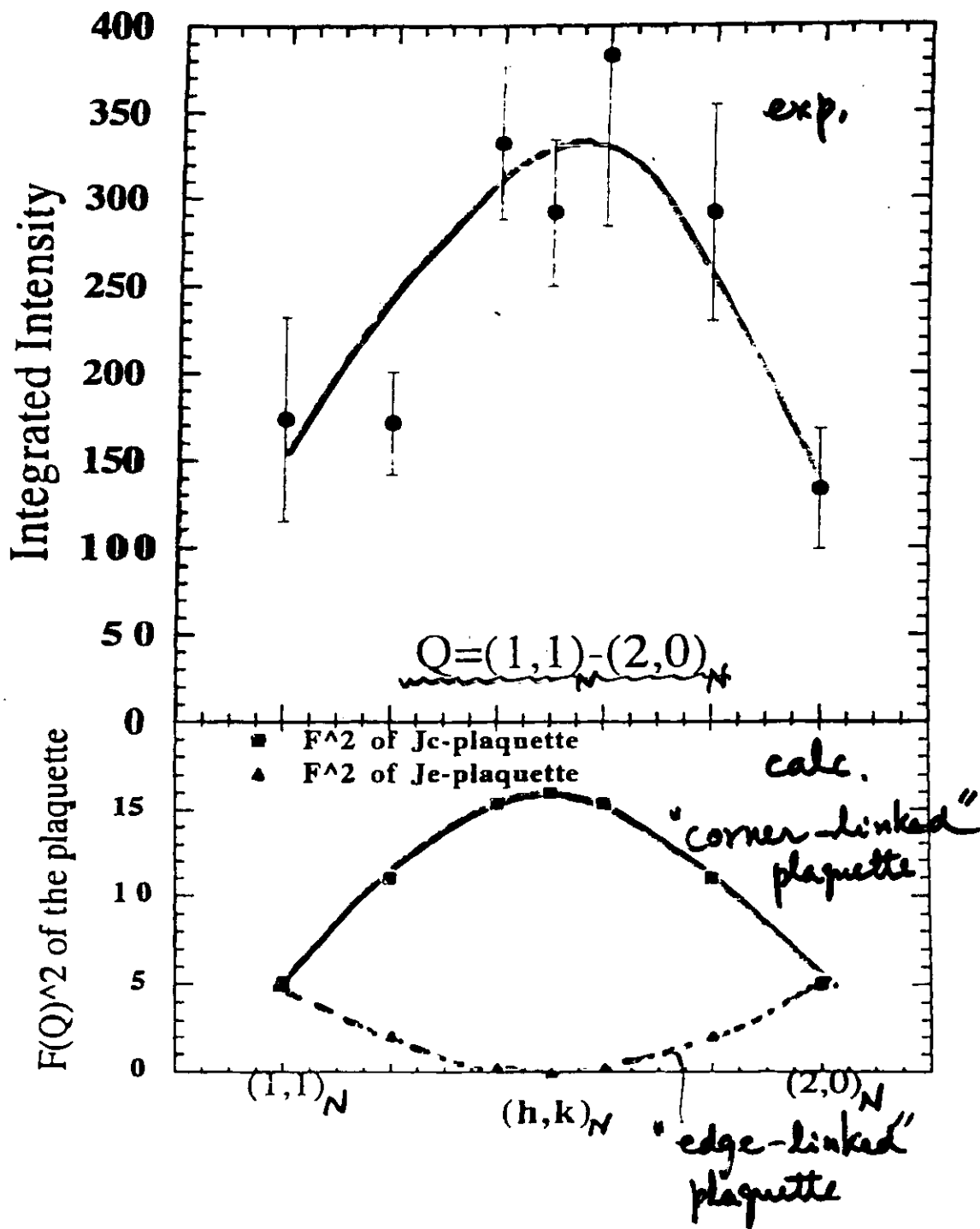
CaV<sub>2</sub>O<sub>7</sub> ( $h, 1, 0$ )

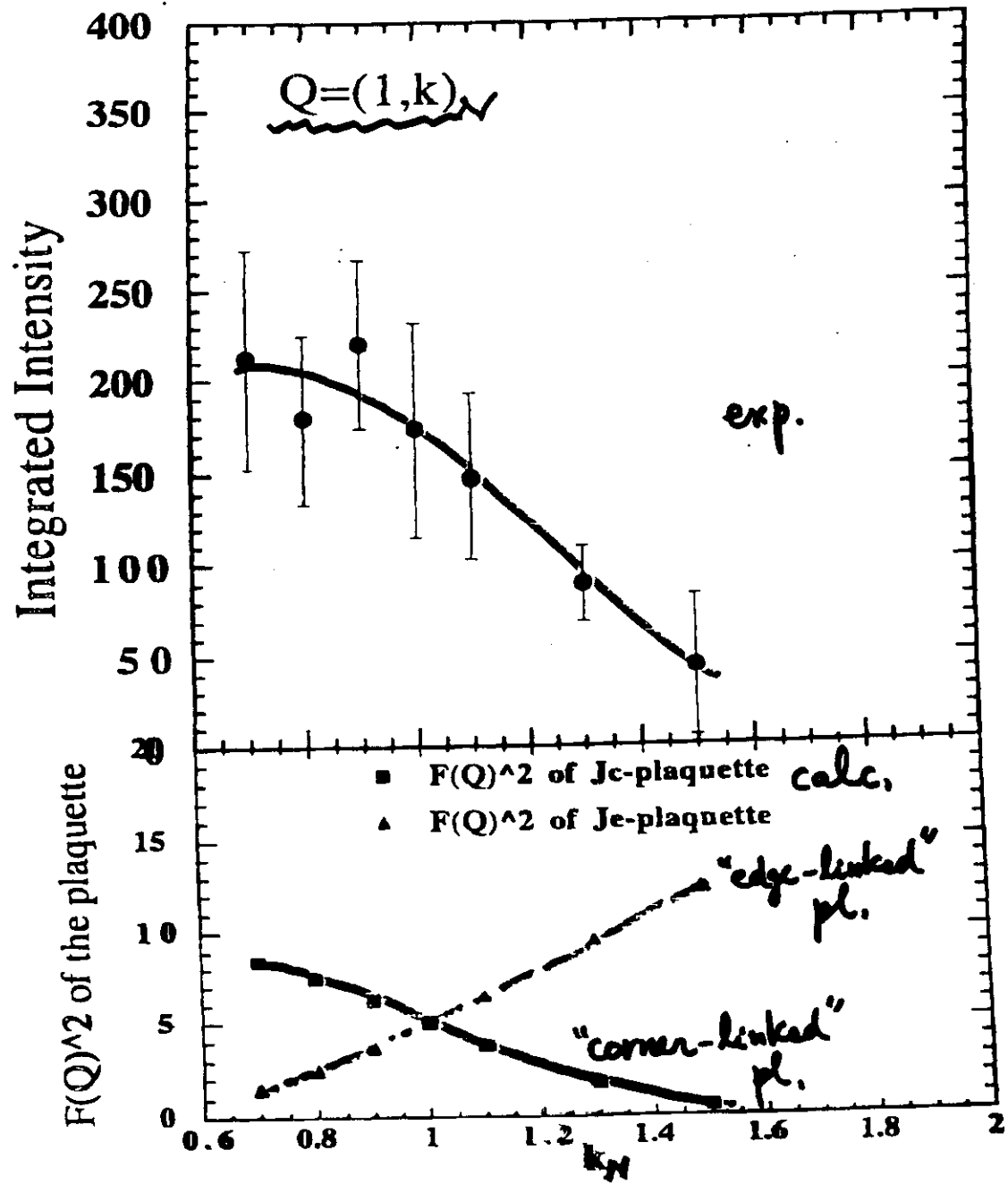
$(-2-\delta, \delta, 0)$

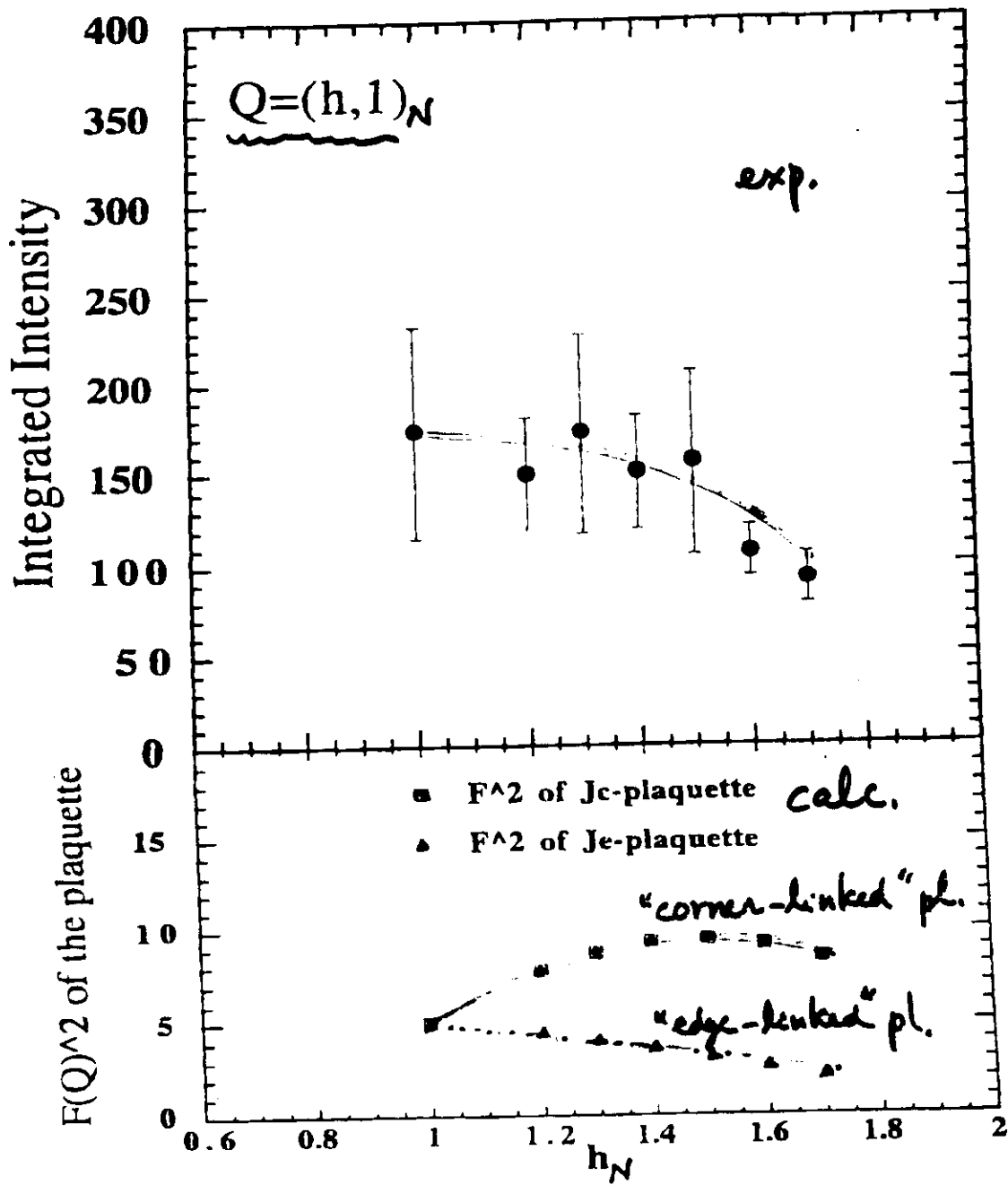




# Structure factor of neutron scattering





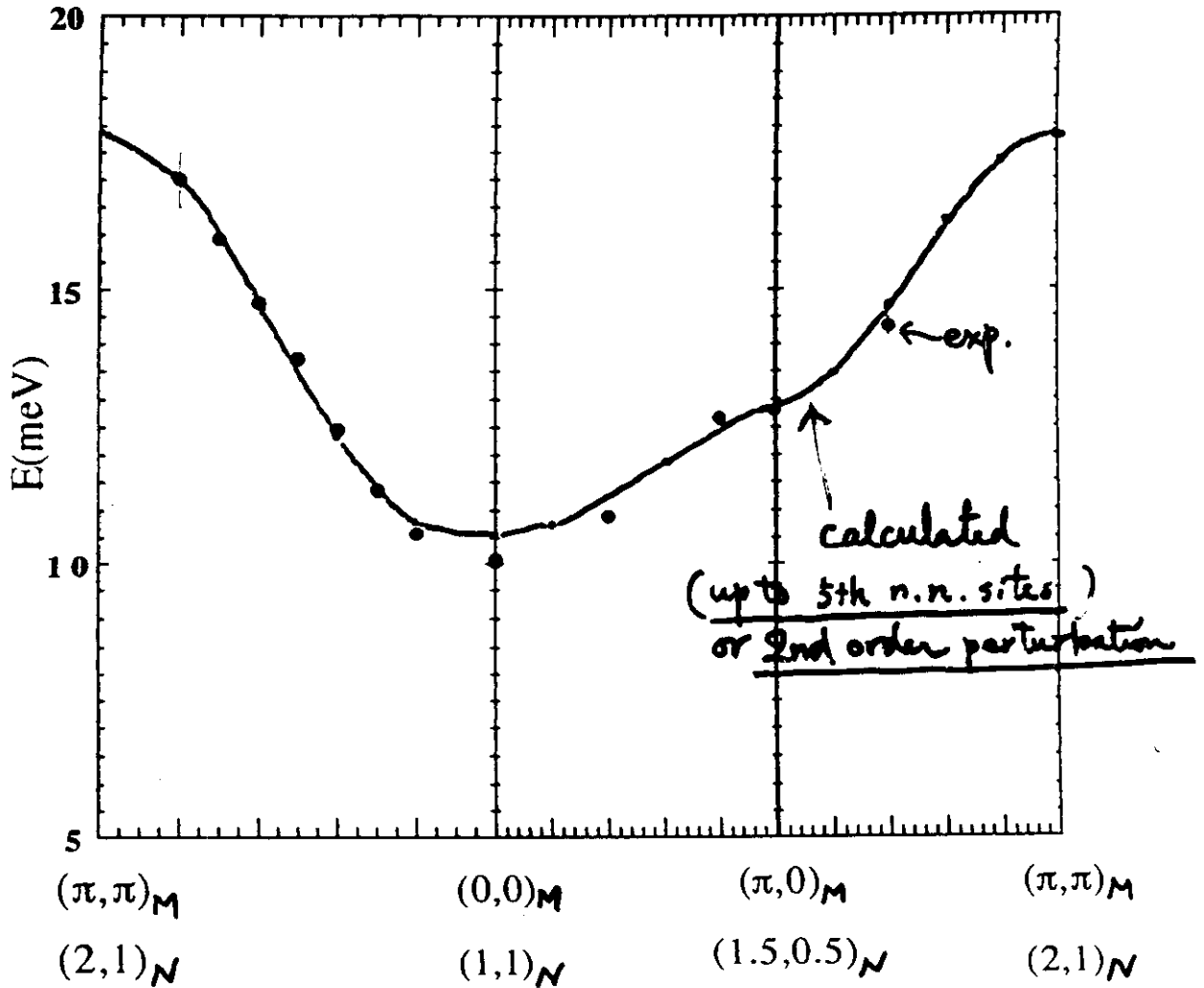


discrepancy  
from higher order effect ?

$$\begin{aligned} \varepsilon_{\frac{1}{2}}(k) - \varepsilon_{\frac{3}{2}} = & J_c + \frac{J_c'}{3} (\cos k_x a + \cos k_y a) + \left(-\frac{J_c}{3} + \frac{J_e'}{6}\right) \times 4 \cos \frac{k_x a}{2} \cos \frac{k_y a}{2} \\ & + \dots \end{aligned}$$

four parameters  $J_c, J_c'$   
 $J_e, J_e'$

$\text{CaV}_4\text{O}_9$



$$\begin{pmatrix} J_c & 14.9 \text{ meV} \\ J_c' & 1.3 \\ J_e = J_e' & 5.9 \end{pmatrix}$$

$$\begin{pmatrix} J_c & 13.4 \\ J_c' & 1.5 \\ J_e & 1.3 \\ J_e' & -3.7 \end{pmatrix}$$

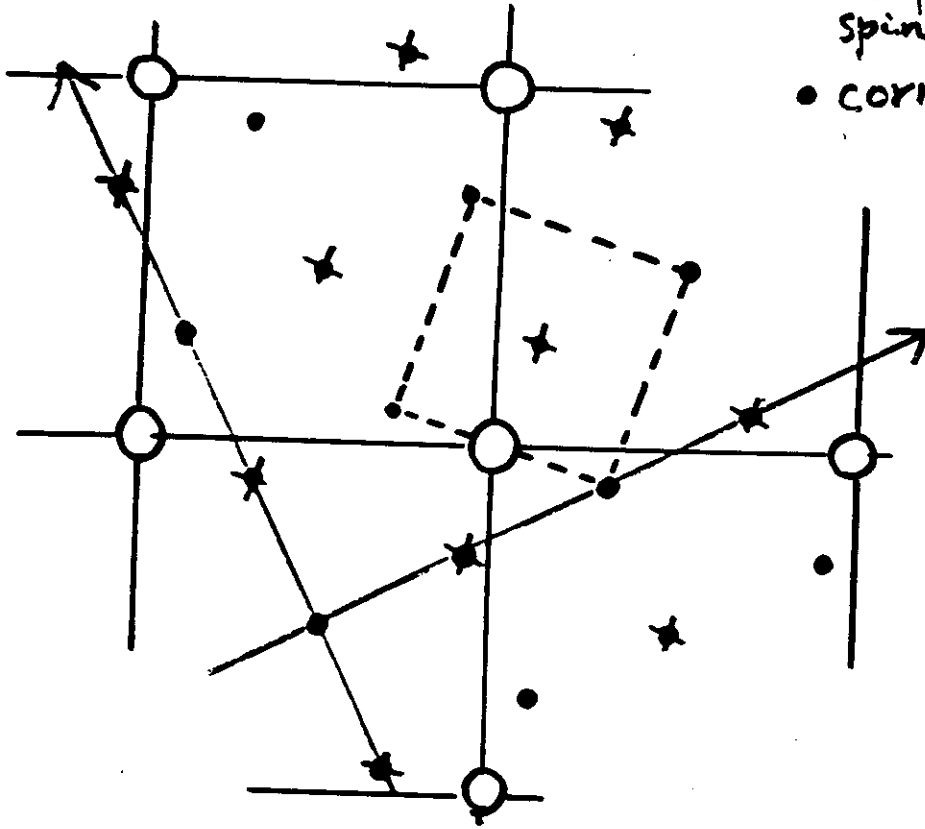
K. Kodama, H. Harashina, M.S.

# CASE III

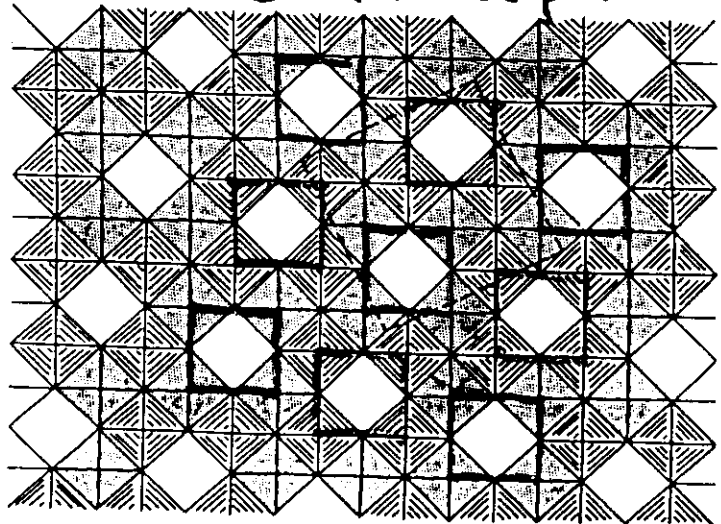
← • mag. unit cell  $\frac{a}{\sqrt{2}} \times \frac{a}{\sqrt{2}}$

• in-phase interplaquette spin correlation

• corner-linked plaquette

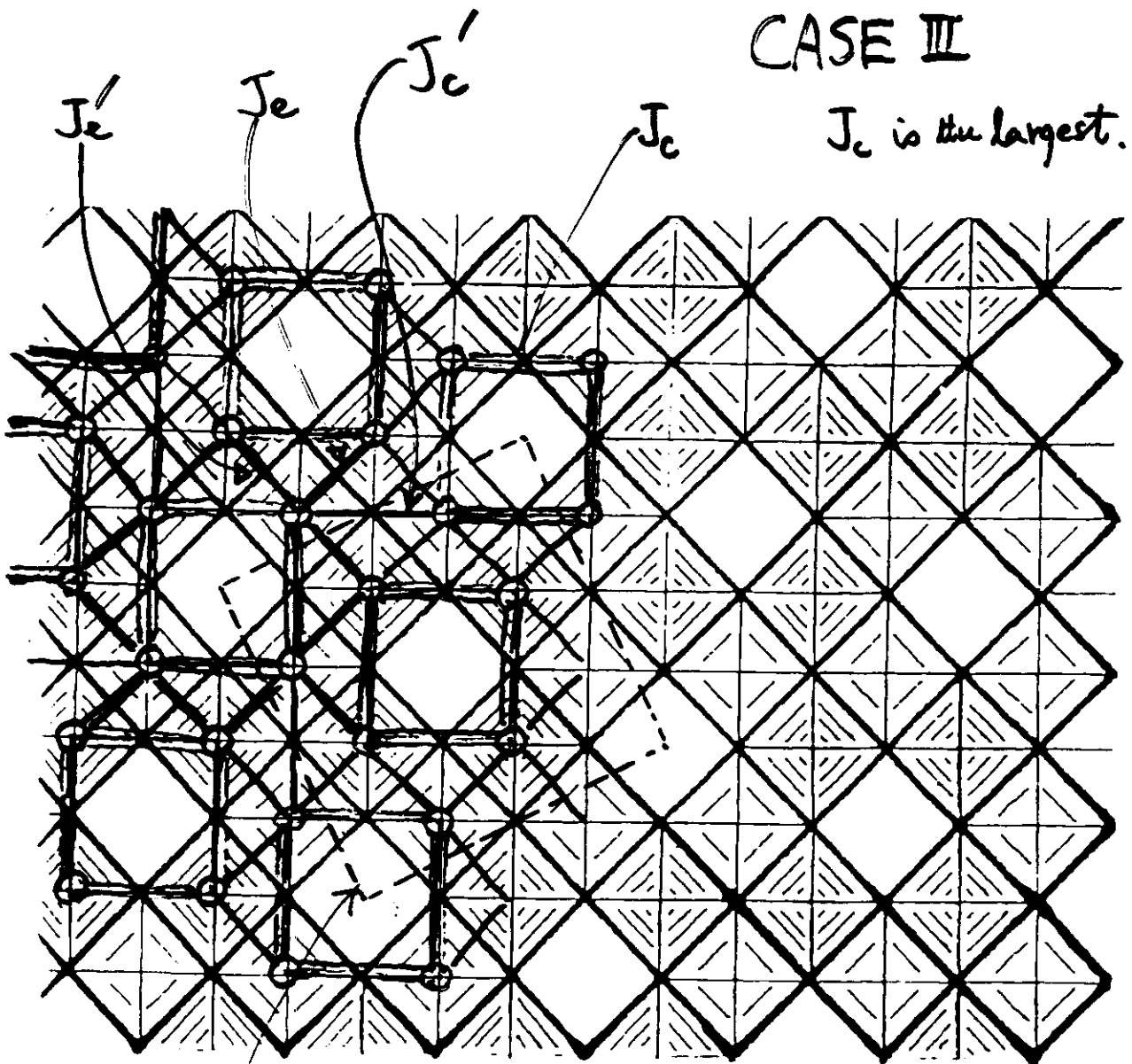


"corner-linked" pl.



# CASE III

$J_c$  is the largest.

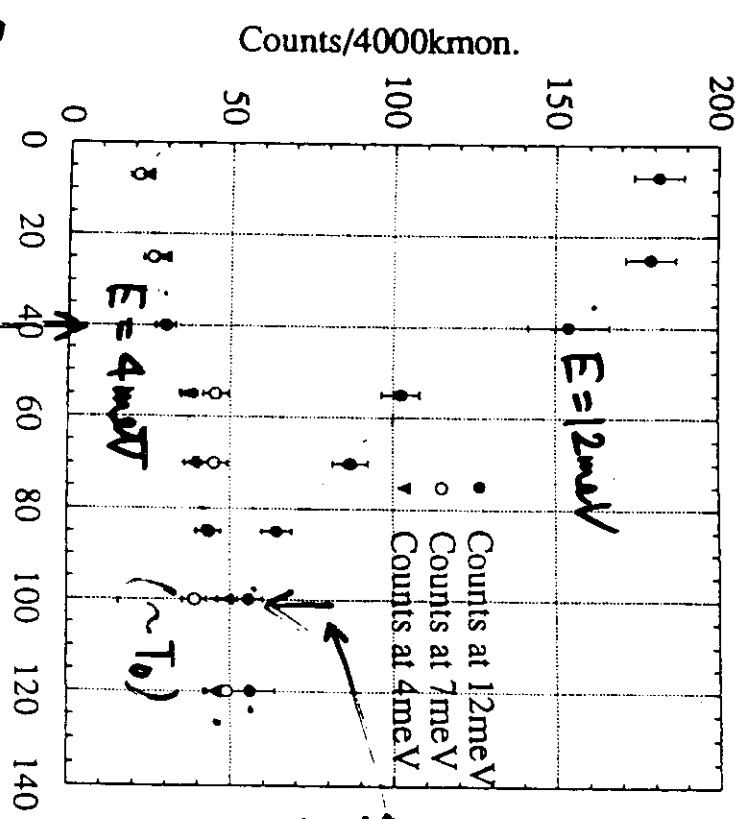
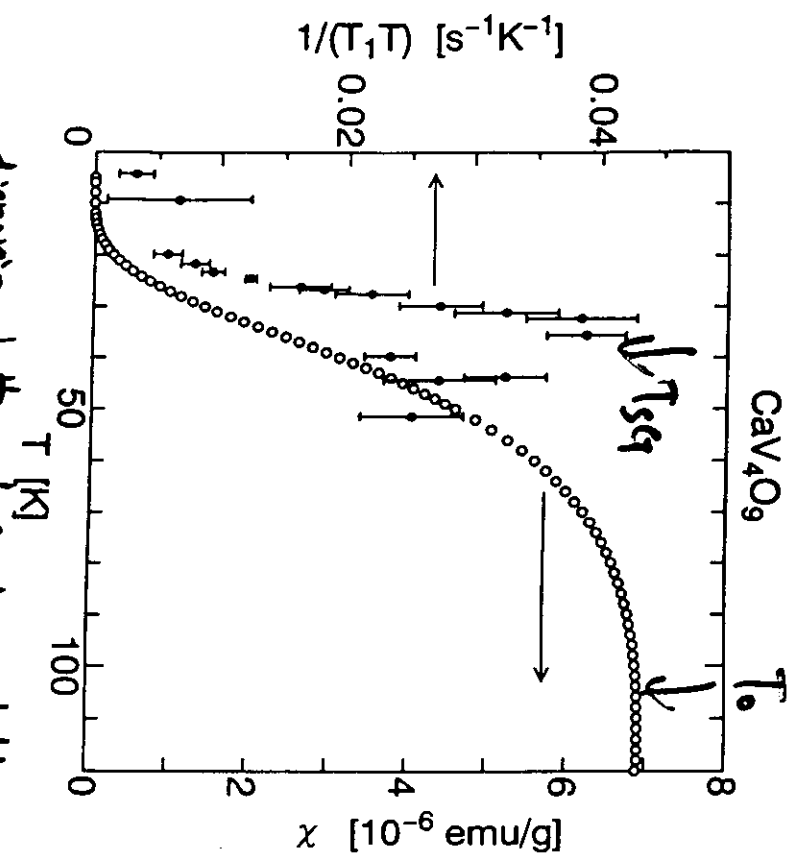


nuclear unit cell

Physical picture of spin gap phase

$J_e$	$o \cdots o$	(x2)
$J_e'$	$\begin{pmatrix} o \cdots o \\ o \cdots o \end{pmatrix}$	(x4)
$J_c'$	$\begin{pmatrix} o \cdots o \\ o \cdots o \end{pmatrix}$	(x4)

neutron scatt. intensity  
 $\chi(q=0)$   
 $(Q = 1.3 \text{ \AA}^{-1})$

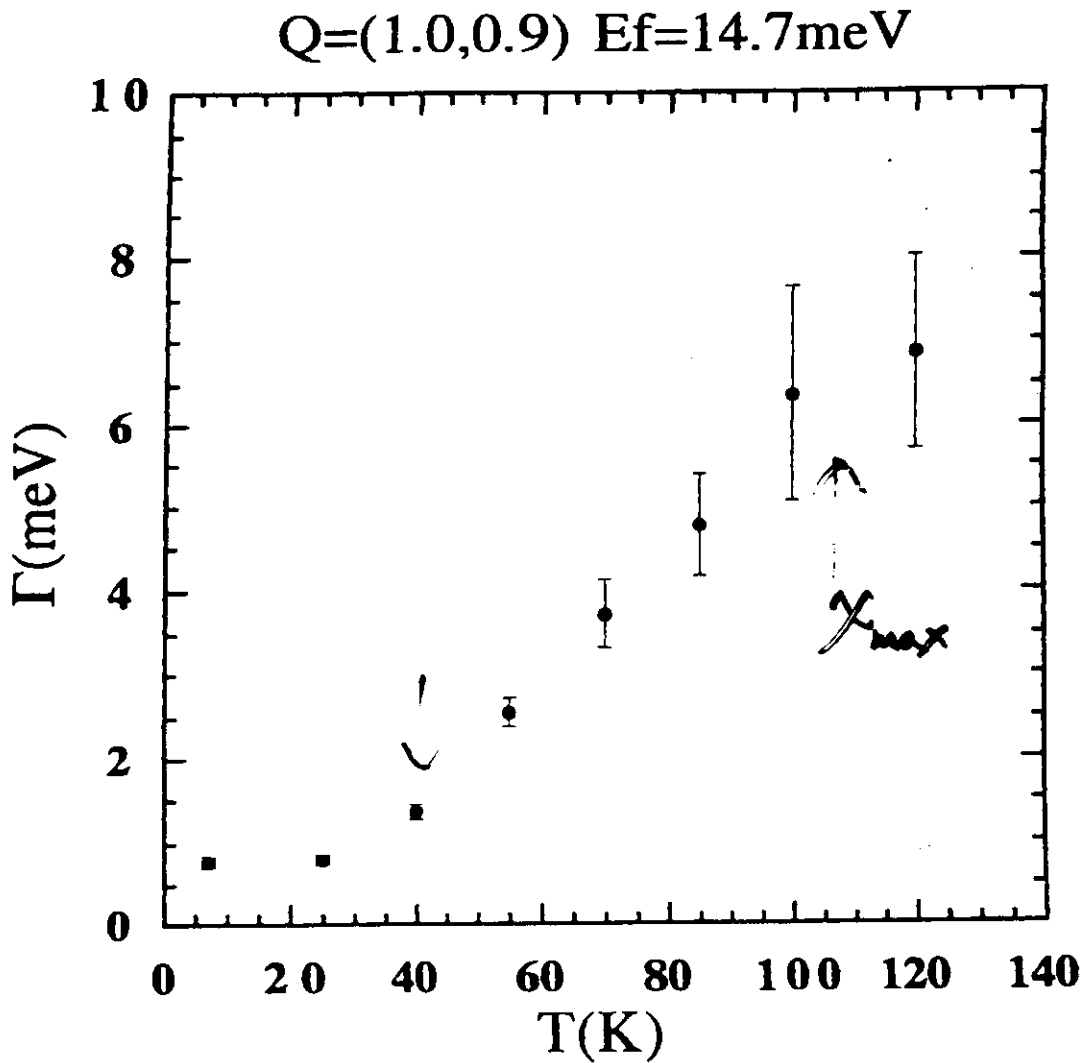


dispersion bottom of  $s \rightarrow z$  excitation at  $P$ .  
 $T_{\text{dep.}}$  of  $1/T_1T$  is dominated by the mag. behavior  
 at  $q=0$ , at least at low  $T$ .

$$\left\{ \begin{array}{l} \chi_{\text{spin}} \\ 1/T_1T \end{array} \right\} \propto e^{-\beta A} \quad (\text{at low } T)$$

$T_{\text{sq}} < T_c$  ?

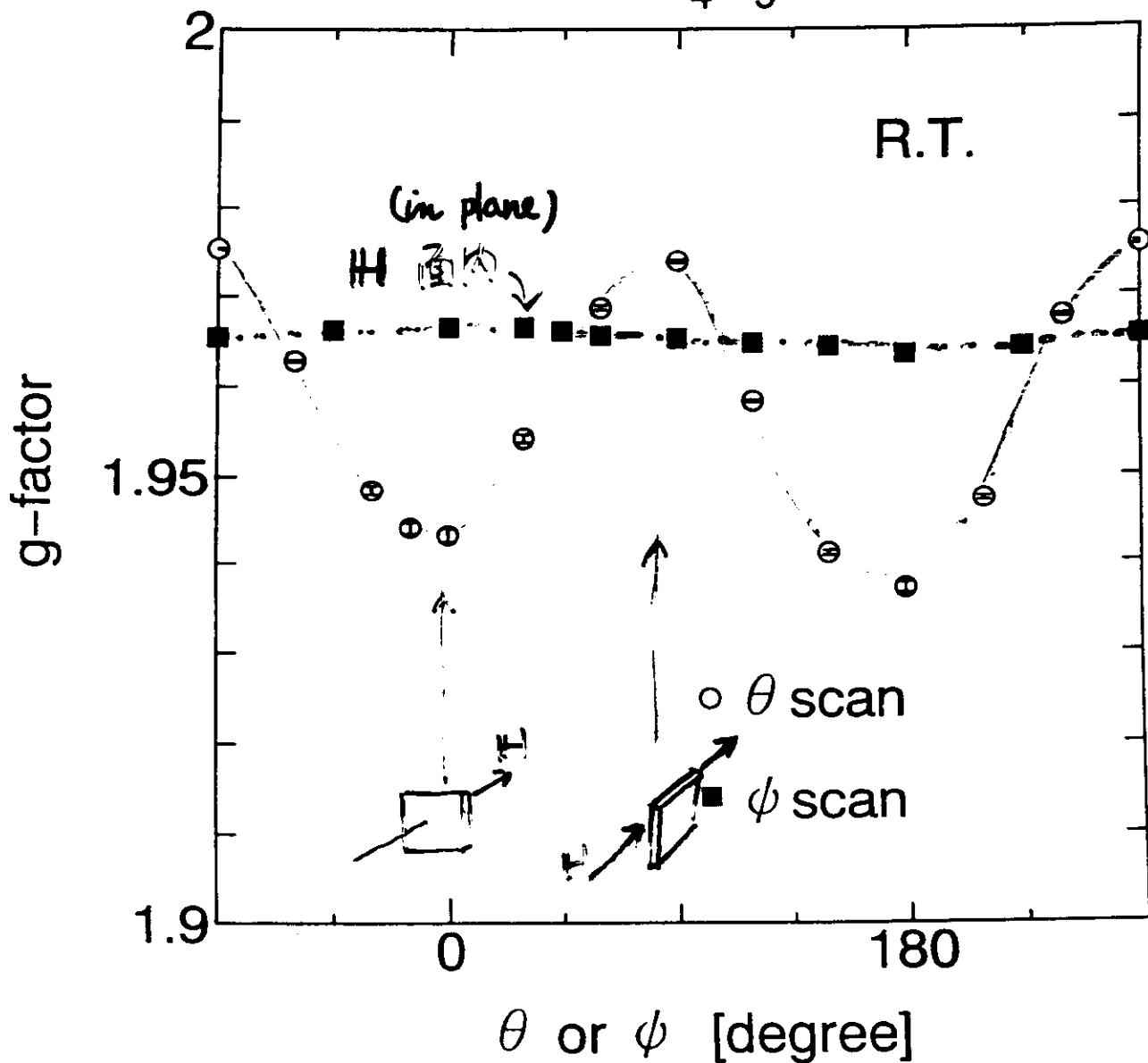
Width of the singlet  $\rightarrow$  triplet excitation  
(at  $k_0$ )



ESR

anisotropy of  $g$ -factor

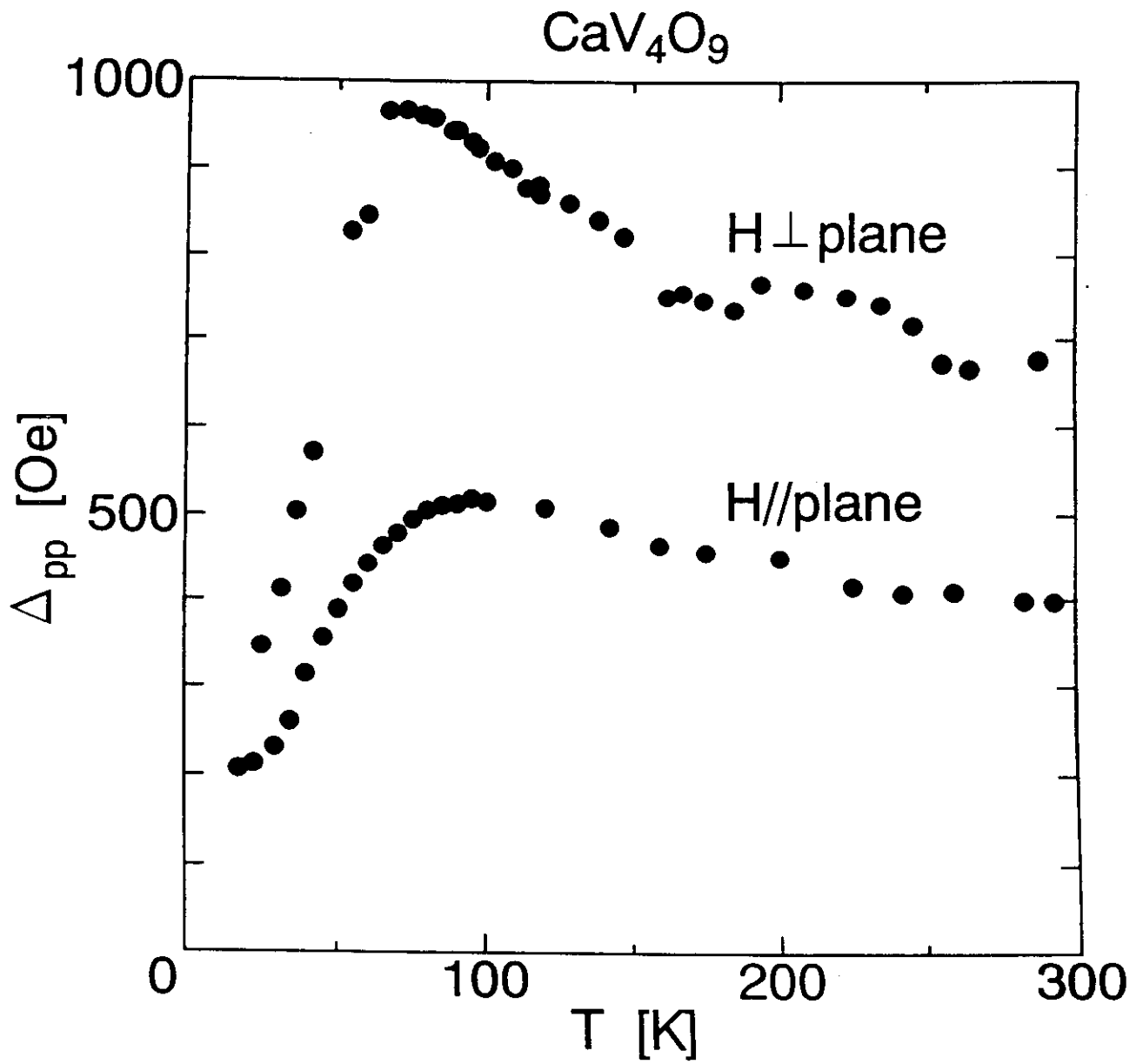
$\text{CaV}_4\text{O}_9$



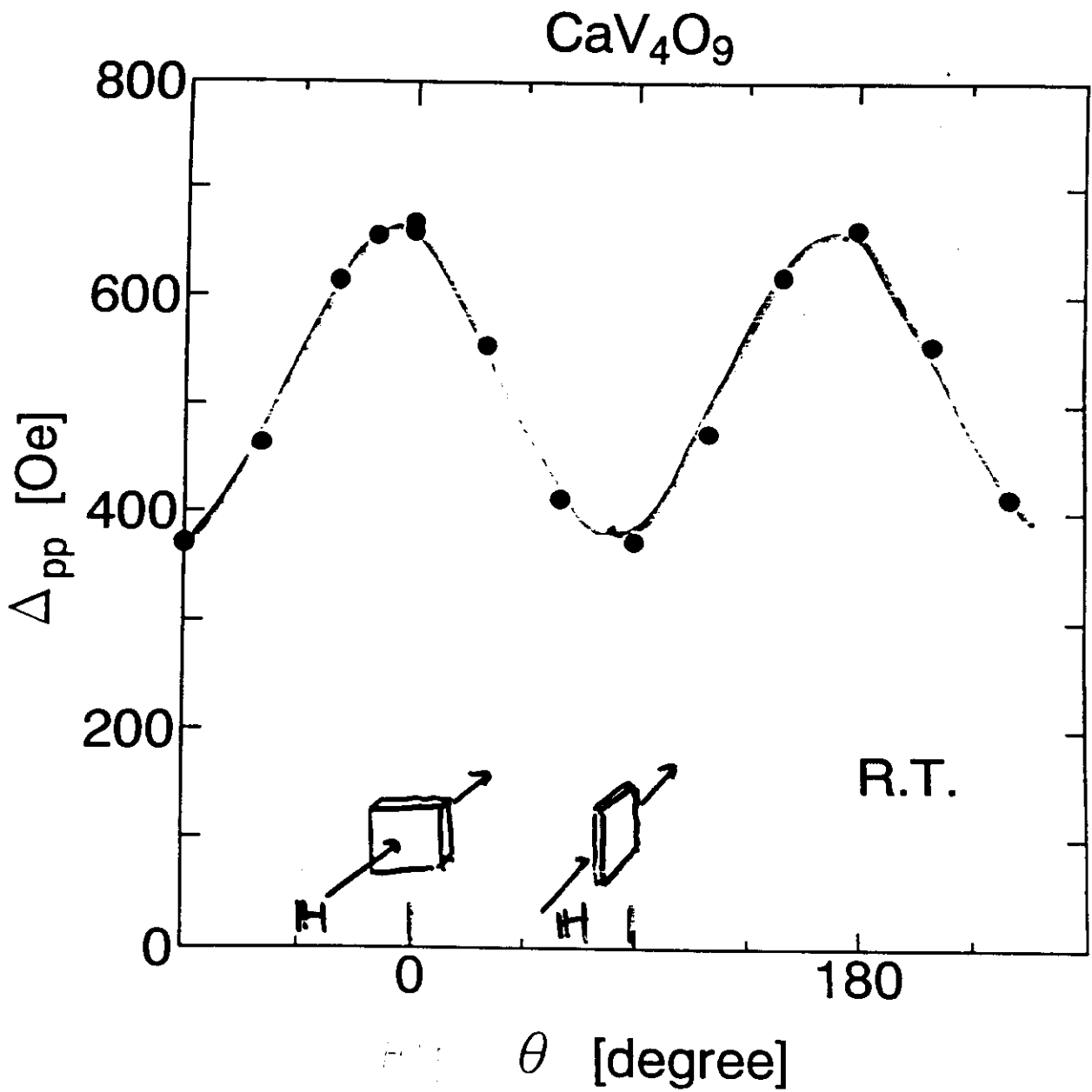
In what orbit does the electron exist ?

S. Taniguchi et al.

ESR width



anisotropy of ESR width



$\theta$ : angle between H and C.

# Summary

$\text{CaV}_4\text{O}_9$

## 2D SPIN GAP SYSTEM

$$\Delta \sim 10 \text{ meV}$$

"corner-linked" plaquette  
observation of the dispersion of  
magnetic excitations



dispersion bottom at  $\Gamma$   
(different from the case of high- $T_c$ )

A physical picture of the spin gap phase  
has been presented.

