科研費
KAKENHI

人をつくり、時代を拓く。
福岡大學

Theory of absorption in the Shastry-Sutherland material $SrCu_2(BO_3)_2$

Shin Miyahara (Fukuoka University, Japan)

Acknowledgement
Nobuo Furukawa (Aoyama Gakuin University, Japan)
Isao Maruyama (Fukuoka Institute of Technology, Japan)

Outline

1. Introduction
2. Frustrated magnetism $SrCu_2(BO_3)_2$
3. Dynamical magnetoelectric effects in $SrCu_2(BO_3)_2$
4. Conclusion

Magnetoelectric(ME) effects

conventional materials

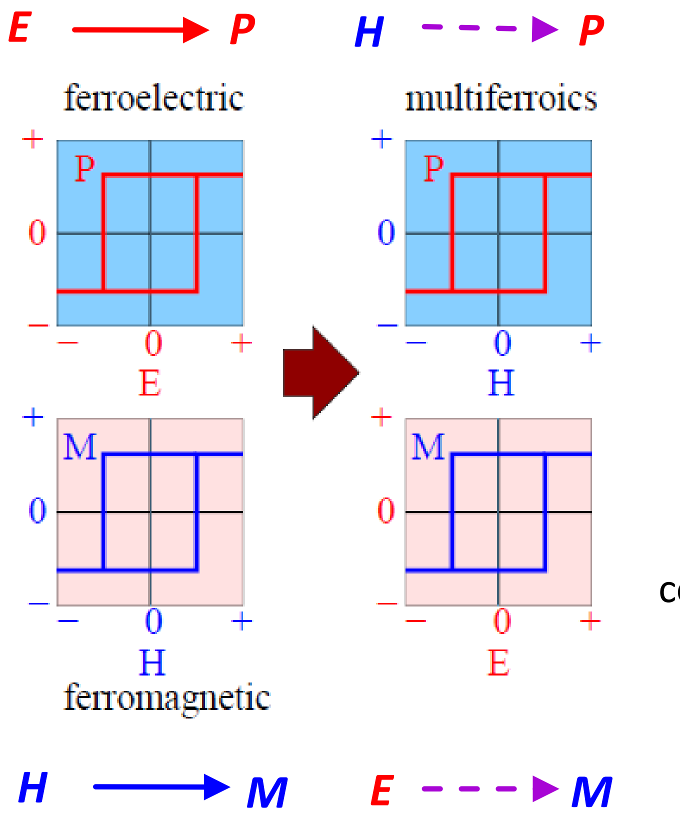
P can be tuned by electric field ***E***
M can be tuned by magnetic field ***H***

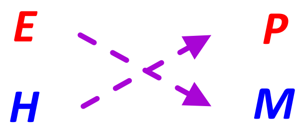
multiferroics
coexistence of multi (anti)ferroic orders

magnetoelectric multiferroics
coexistence of ***P*** and ***M***

P can be tuned by magnetic field ***H***
M can be tuned by electric field ***E***

ferroelectricity ***P***, ferromagnetism ***M***





cross correlation

coupling between ***P*** and ***M***
e.g. spin current mechanism

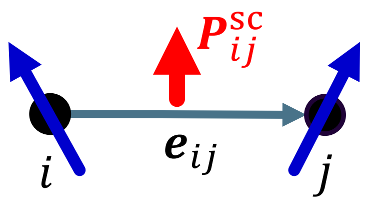
$$P_{ij}^{sc} = d\boldsymbol{e}_{ij} \times (\boldsymbol{S}_i \times \boldsymbol{S}_j)$$

Electric polarization due to spin current mechanism

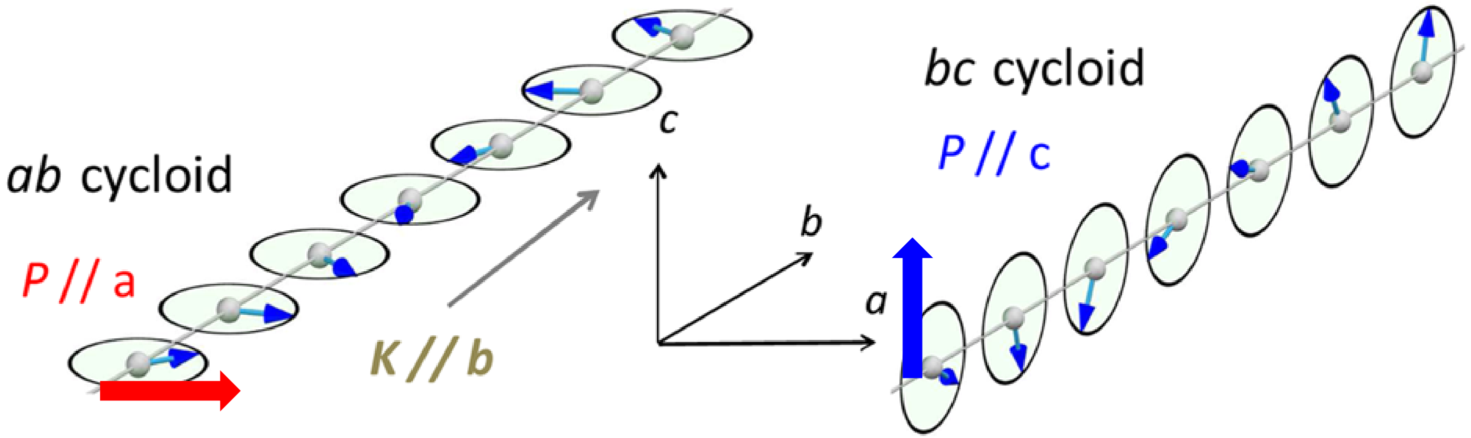
Electric polarization due to spin-current (inverse DM interaction) mechanism

$$\boldsymbol{P}_{ij}^{sc} = d\boldsymbol{e}_{ij} \times (\boldsymbol{S}_i \times \boldsymbol{S}_j)$$

coupling between ***P*** and spin-pair



H. Katsura *et al*., Phys. Rev. Lett. **95** 057205 (2005),
M. Mostovoy, Phys. Rev. Lett. **96** 067601 (2006)
I.A. Sergienko and E. Dagotto, Phys. Rev. Lett. **96** 067601 (2006)



cycloidal plane flops by *H* // *c*
(*ab*-cycloid➔*bc*-cycloid)

electric polarization flop
(*P* // *a* ➔ *P* // *c*)

Dynamical magentoelectric effects

dynamical ME effect:
cross correlation by time-dependent fields

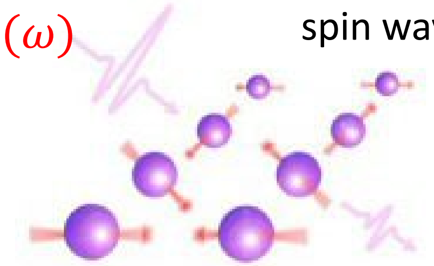
$\boldsymbol{M}(\boldsymbol{\omega})$ is induced by electric field $E(\omega)$
$\boldsymbol{P}(\boldsymbol{\omega})$ is induced by magnetic field $H(\omega)$

$E(\omega) \rightarrow \boldsymbol{P}(\boldsymbol{\omega})$

$H(\omega) \rightarrow \boldsymbol{M}(\boldsymbol{\omega})$

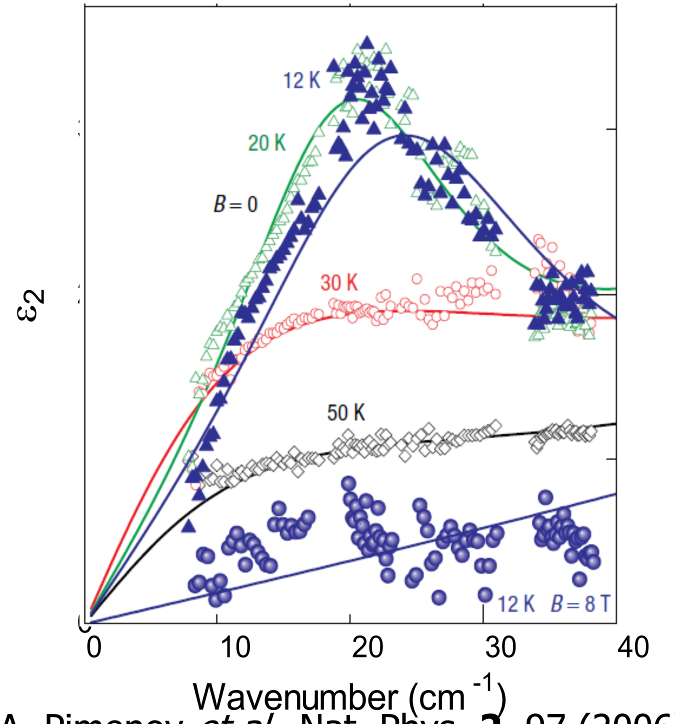
electromagnon
electro-active magnon
(mostly in GHz～THz region)

$E(\omega)$ spin wave



N. Kida *et al.*, Phys. Rev. B **78**, 104414 (2008)

electromagnon in *R*MnO_3

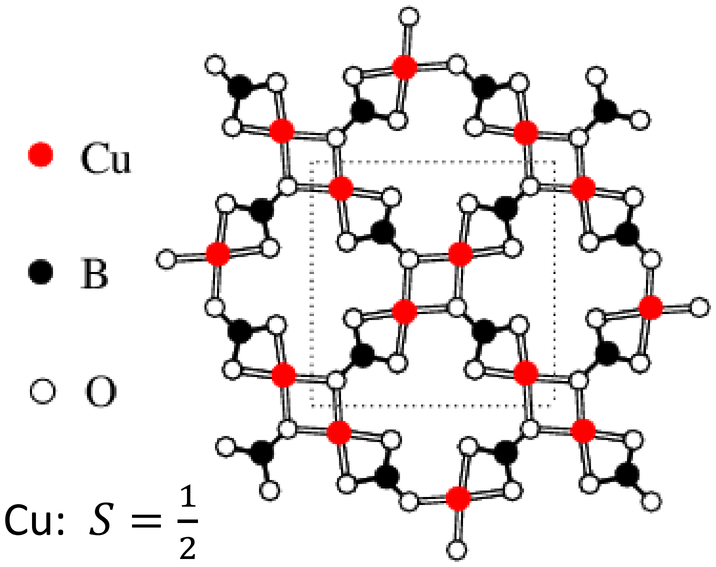


A. Pimenov *et al.*, Nat. Phys. **2**, 97 (2006)

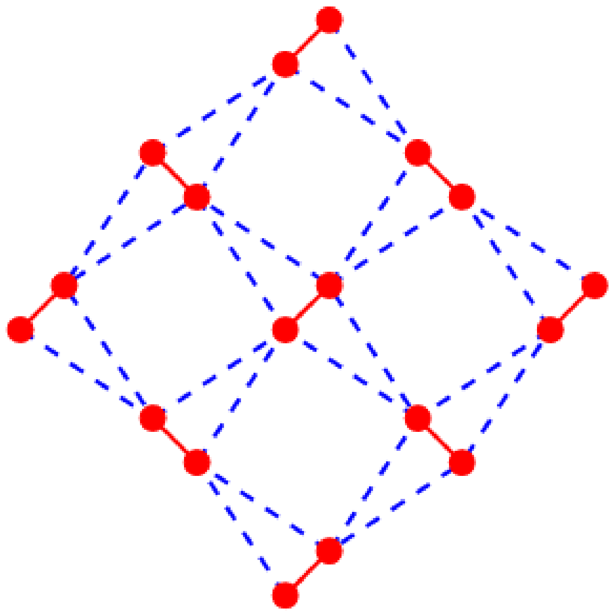
exploring other cases of the electro-active magnetic excitation

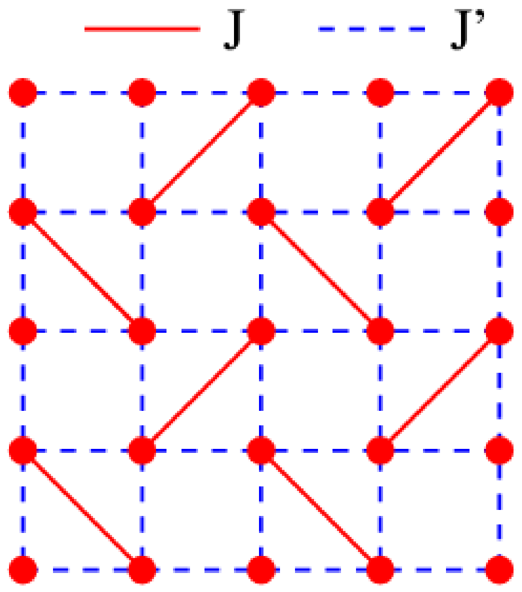
spin gap system $SrCu_2(BO_3)_2$

Two-dimensional spin gap system $SrCu_2(BO_3)_2$



Cu: $S = \frac{1}{2}$





$Cu(BO_3)$ plane

orthogonal dimer model

Shastry-Sutherland model

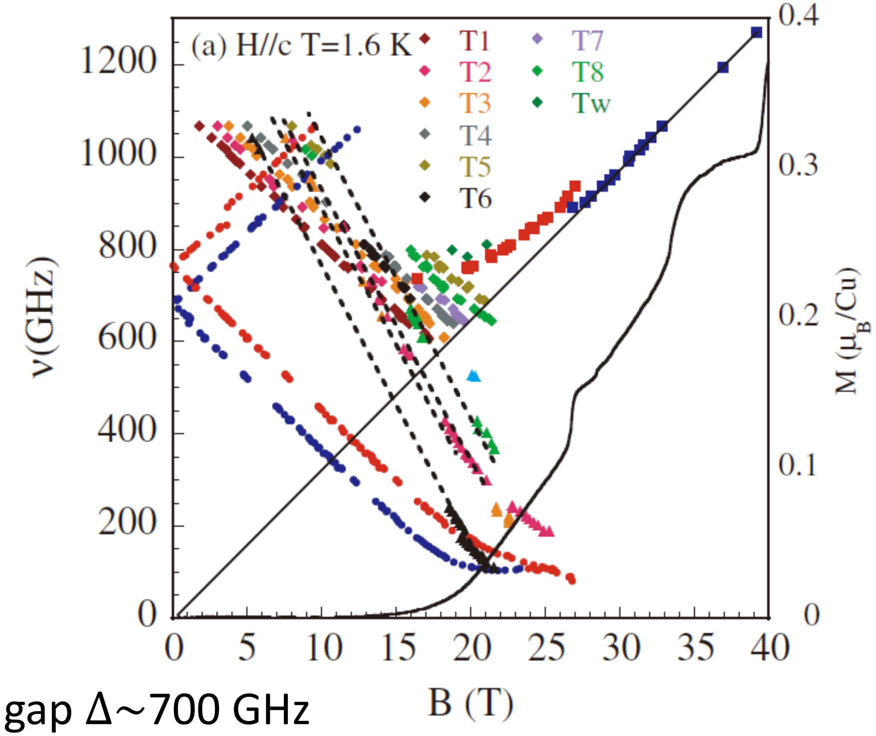
spin singlet ground state
spin gap $\Delta \sim 35$K, 730 GHz
magnetization plateaux

exact dimer singlet states

B.S. Shastry and B. Sutherland, Physica B **108** 1069 (1981)
H. Kageyama *et al.*, Phys. Rev. Lett. **82** 3168 (1999)
S. Miyahara and K. Ueda, Phys. Rev. Lett. **82** 3701 (1999)

Magnetic features of $SrCu_2(BO_3)_2$

ESR



spin gap Δ~700 GHz

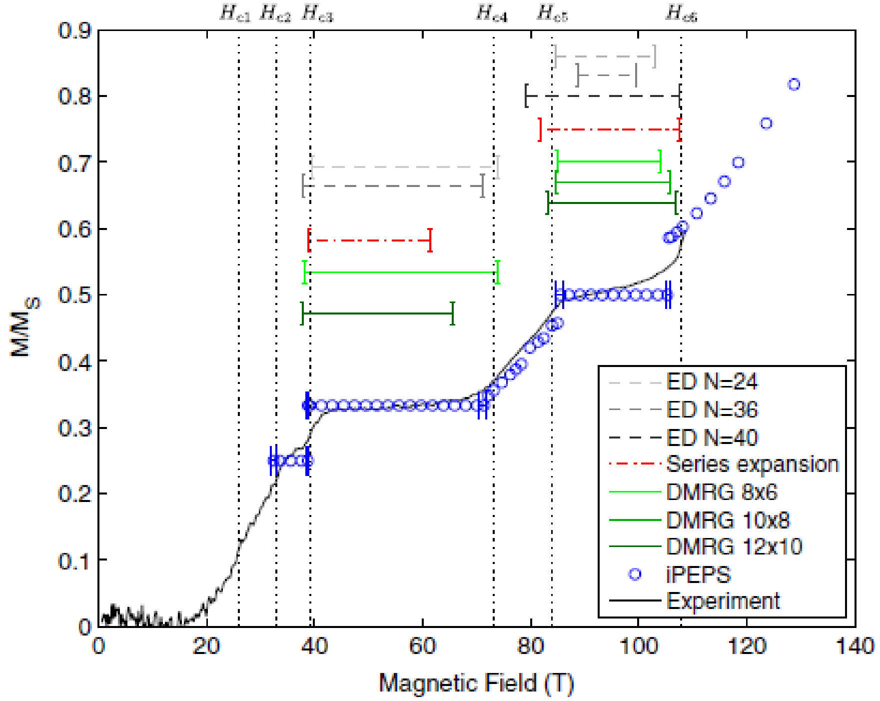
splitting of the triplet excitation due to DM interactions

H. Nojiri *et al*, J. Phys. Soc. Jpn. **68** 2906 (1999) , ibid. **72** 3243 (2003)

T. Rõõm *et al*, Phys. Rev. B **61** 14342 (2000)

O. Cepas *et al*, Phys. Rev. Lett. **87** 167205 (2001)

magnetization plateaux

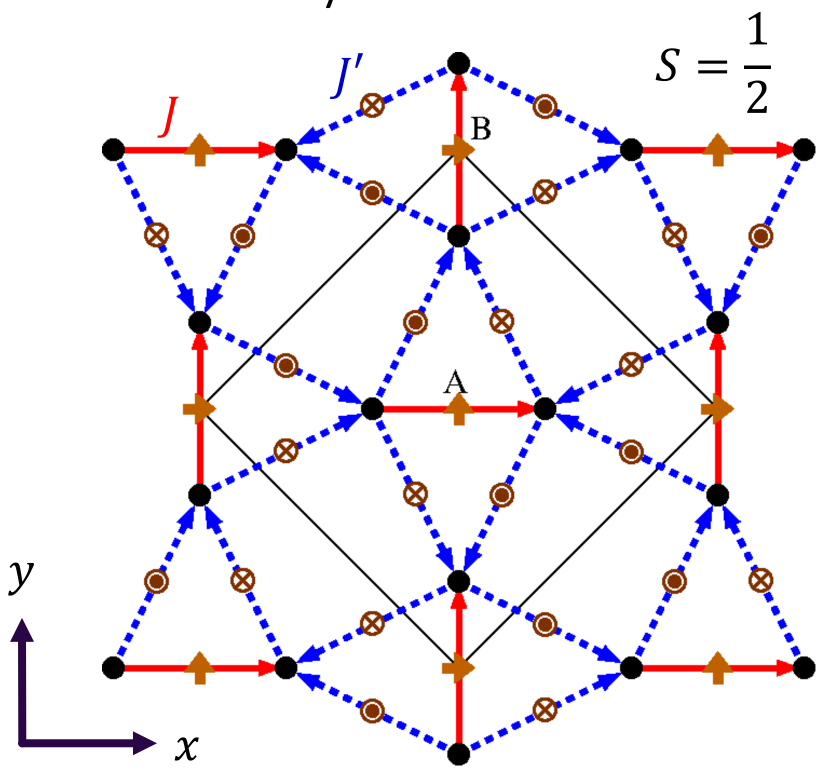


magnetization plateaux at 1/4, 1/3, 1/2

Y.H. Matsuda *et al*, Phys. Rev. Lett. **111** 137204 (2013)

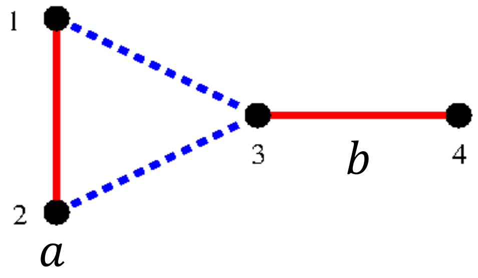
Exact ground state and spin gap of the Shastry-Sutherland model

Shastry-Sutherland model



$$\hat{H} = J \sum_{\text{n.n.}} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + J' \sum_{\text{n.n.n.}} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

ground state



$$h'_{ab} = \boldsymbol{J'S}_1 \cdot \boldsymbol{S}_3 + \boldsymbol{J'S}_2 \cdot \boldsymbol{S}_3 = \boldsymbol{J'}(\boldsymbol{S}_1 + \boldsymbol{S}_2) \cdot \boldsymbol{S}_3$$

$$h'_{ab}|s\rangle_a |s\rangle_b = 0 \qquad |s\rangle_\alpha = \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle_\alpha - |\downarrow\uparrow\rangle_\alpha\right)$$

product of dimer singlet states

$$|\mathrm{GS}\rangle = \prod_\alpha \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle_\alpha - |\downarrow\uparrow\rangle_\alpha\right)$$

exact ground state when $\frac{J'}{J} < 0.675$

B.S. Shastry and B. Sutherland, Physica B **108** 1069 (1981)

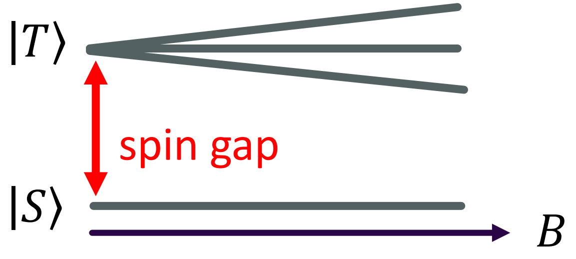
A. Koga and N. Kawakami, PRL **84** 4461 (2000), F. Corboz and F. Mila, PRB **87** 115144 (2013)

spin gap

one of the dimer singlet ➡ triplet

$$|t_1\rangle_\alpha = |\uparrow\uparrow\rangle_\alpha |t_0\rangle_\alpha = \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle_\alpha + |\downarrow\uparrow\rangle_\alpha\right) \; |t_{-1}\rangle_\alpha = |\downarrow\downarrow\rangle_\alpha$$

3rd order of perturbation $\frac{\Delta}{J} = 1 - \left(\frac{J'}{J}\right)^2 - \frac{1}{2}\left(\frac{J'}{J}\right)^3$



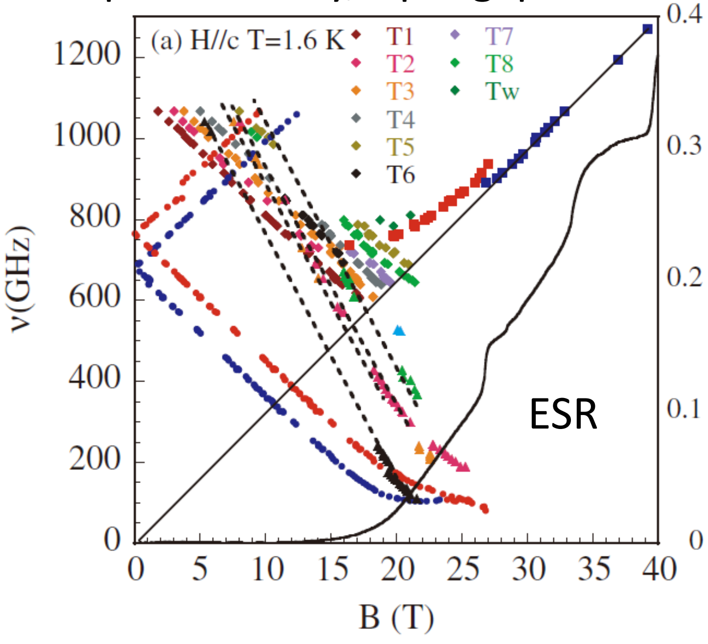
Absorption of spin excitation in $SrCu_2(BO_3)_2$

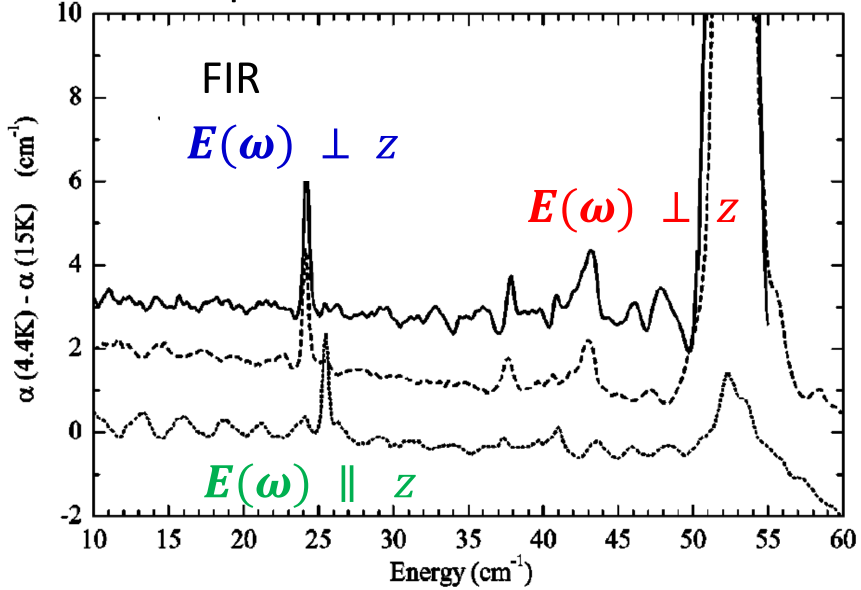
spin gap excitation

transition from singlet g.s. to triplet excitations   forbidden transition in Heisenberg model

c.f. Anisotropic terms induce magneto active mode.   *e.g.* M. Oshikawa , JPSJ **72** Suppl. B 36 (2003), T. Saka and H. Shiba JPSJ **63** 867 (1994)

Experimentally, spin gap and bound states of triplet excitations have been observed in absorption





electro-active magnetic excitations

spin gap excitation

$24.2\ \mathrm{cm}^{-1}$ $E(\omega) \perp z$

$25.7\ \mathrm{cm}^{-1}$ $E(\omega) \parallel z$

bound states of two triplets

$37.5, 43.0, 52.3, 53.5\ \mathrm{cm}^{-1}$

$E(\omega) \perp z$

H. Nojiri *et al*, J. Phys. Soc. Jpn. **68** 2906 (1999) , ibid. **72** 3243 (2003)

T. Rõõm *et al*, Phys. Rev. B **61** 14342 (2000), ibid **70** 14417 (2004) , O. Cepas *et al*, Phys. Rev. Lett. **87** 167205 (2001)

Purpose : clarify the origin of the spin gap excitation (selection rule of absorption)

Dynamical electric susceptibility

Dynamical electric susceptibility

$$\varepsilon^{\beta}_{\alpha\alpha}(\omega) \propto \left\langle 0 \middle| \hat{P}^{\beta\dagger}_{\alpha} \frac{1}{\omega + E_0 + i\varepsilon - \hat{H}} \hat{P}^{\beta}_{\alpha} \middle| 0 \right\rangle$$

$$\alpha = x, y, z \qquad \beta = \mathrm{AS}, S$$

spin current mechanism

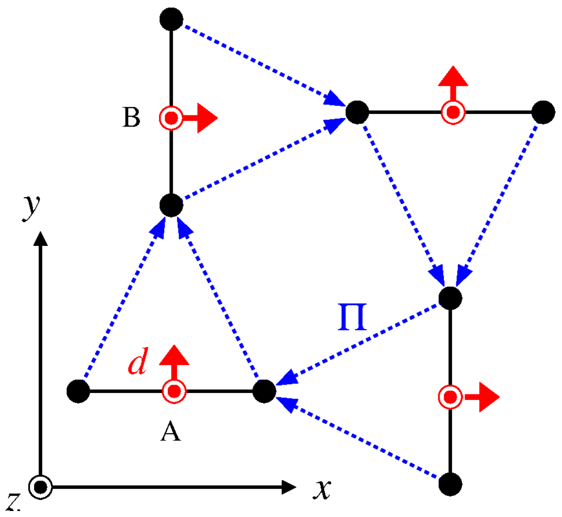
$$\hat{\boldsymbol{P}}^{\mathrm{AS}} = \sum_{\mathrm{n.n.}} d\boldsymbol{e}_{ij} \times (\boldsymbol{S}_i \times \boldsymbol{S}_j)$$

$\boldsymbol{E}(\boldsymbol{\omega}) \perp z$ $\quad$ $\boldsymbol{E}(\boldsymbol{\omega}) \parallel z$

exchange striction mechanism

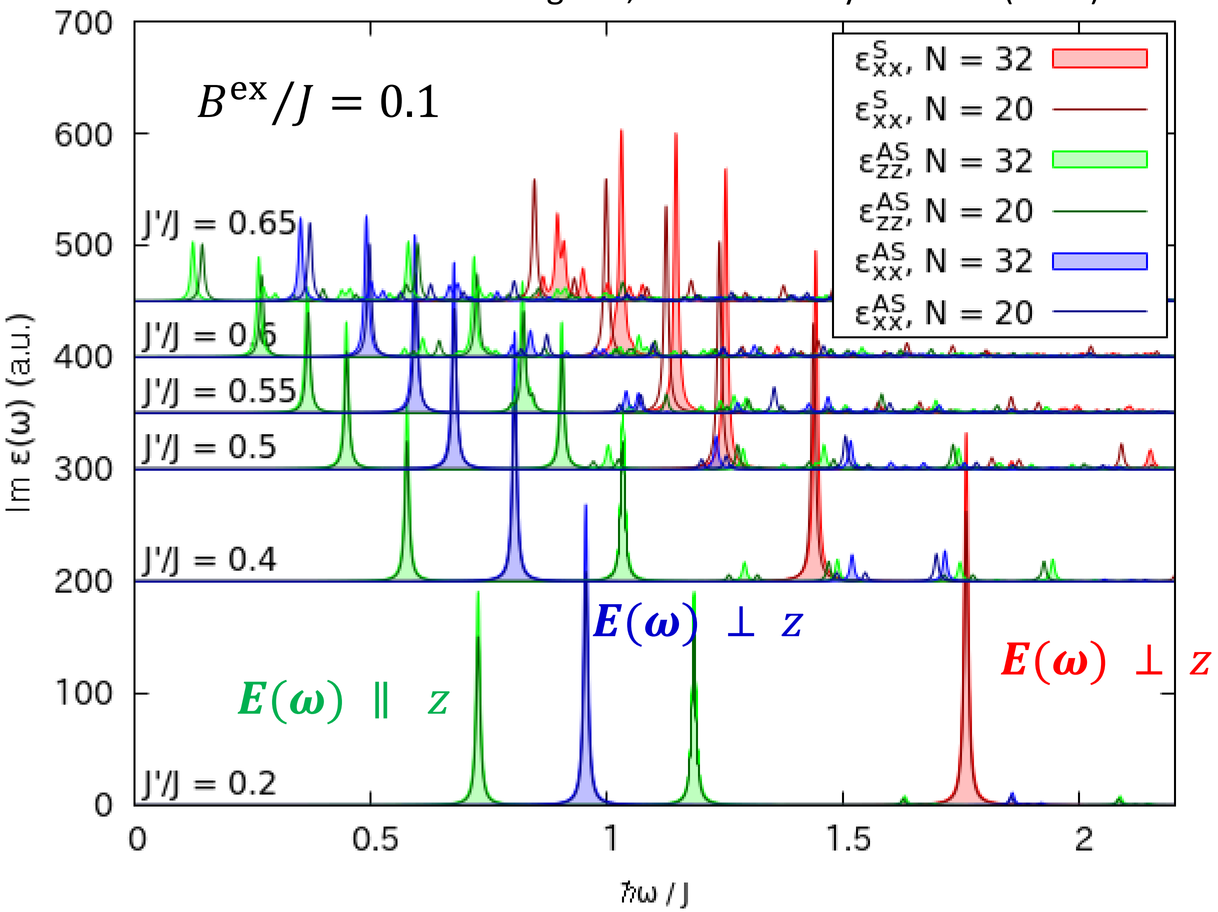
$$\hat{\boldsymbol{P}}^{\mathrm{S}} = \sum_{\mathrm{n.n.n.}} \Pi \boldsymbol{e}_{ij}\, \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

$\boldsymbol{E}(\boldsymbol{\omega}) \perp z$

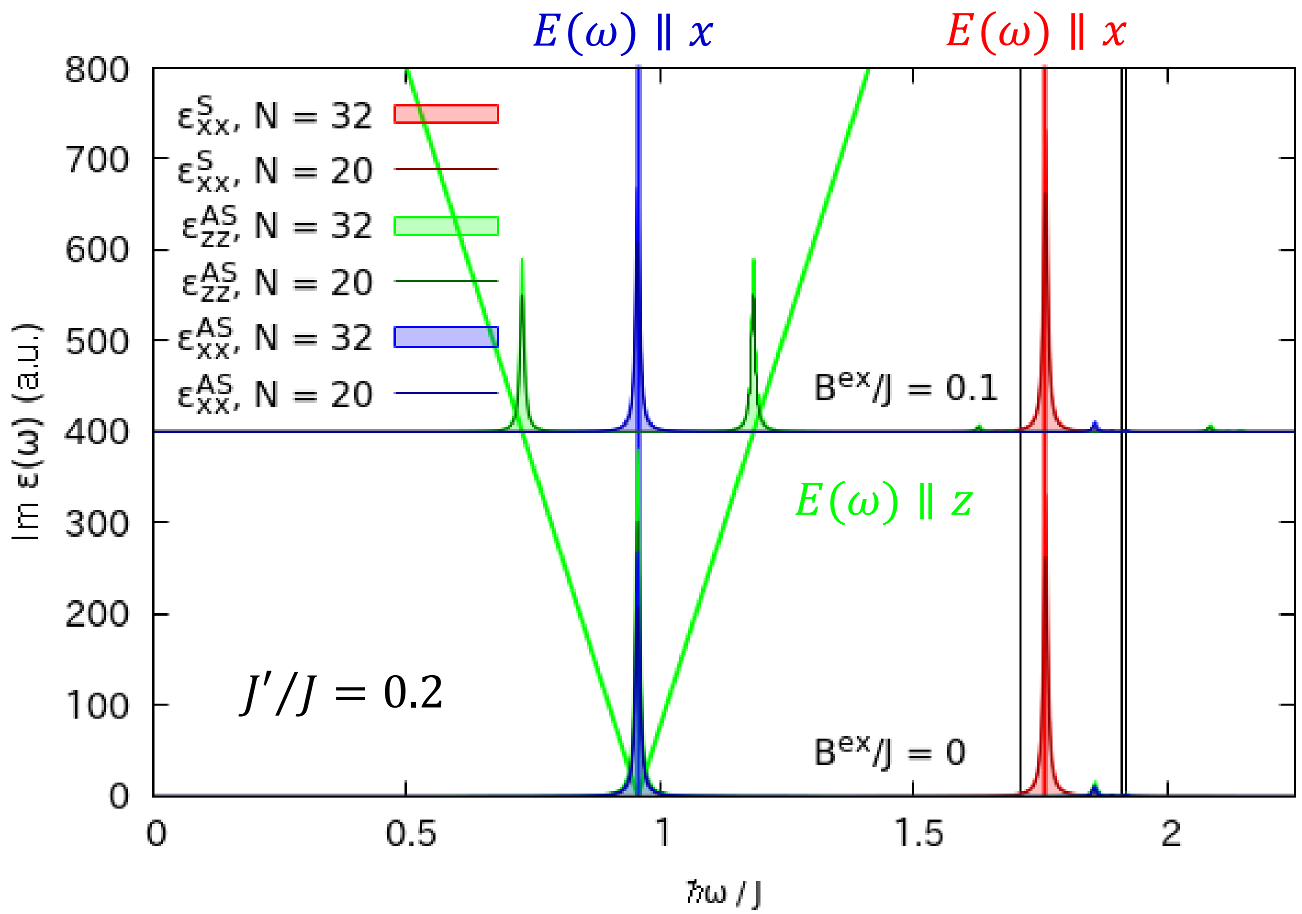


the continued fraction method by using Lanczos method

E. Dagotto, Rev. Mod. Phys. **66** 763 (1994)



Comparison with the perturbation calculation



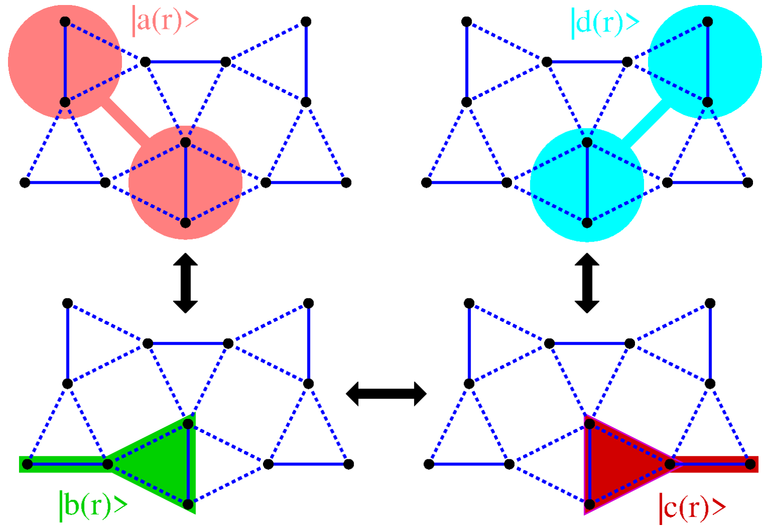
spin gap and $S^T = 0$ bound states are electro active.

3rd order of perturbation

spin gap

$$\frac{\Delta}{J} = 1 - \left(\frac{J'}{J}\right)^2 - \frac{1}{2}\left(\frac{J'}{J}\right)^3$$

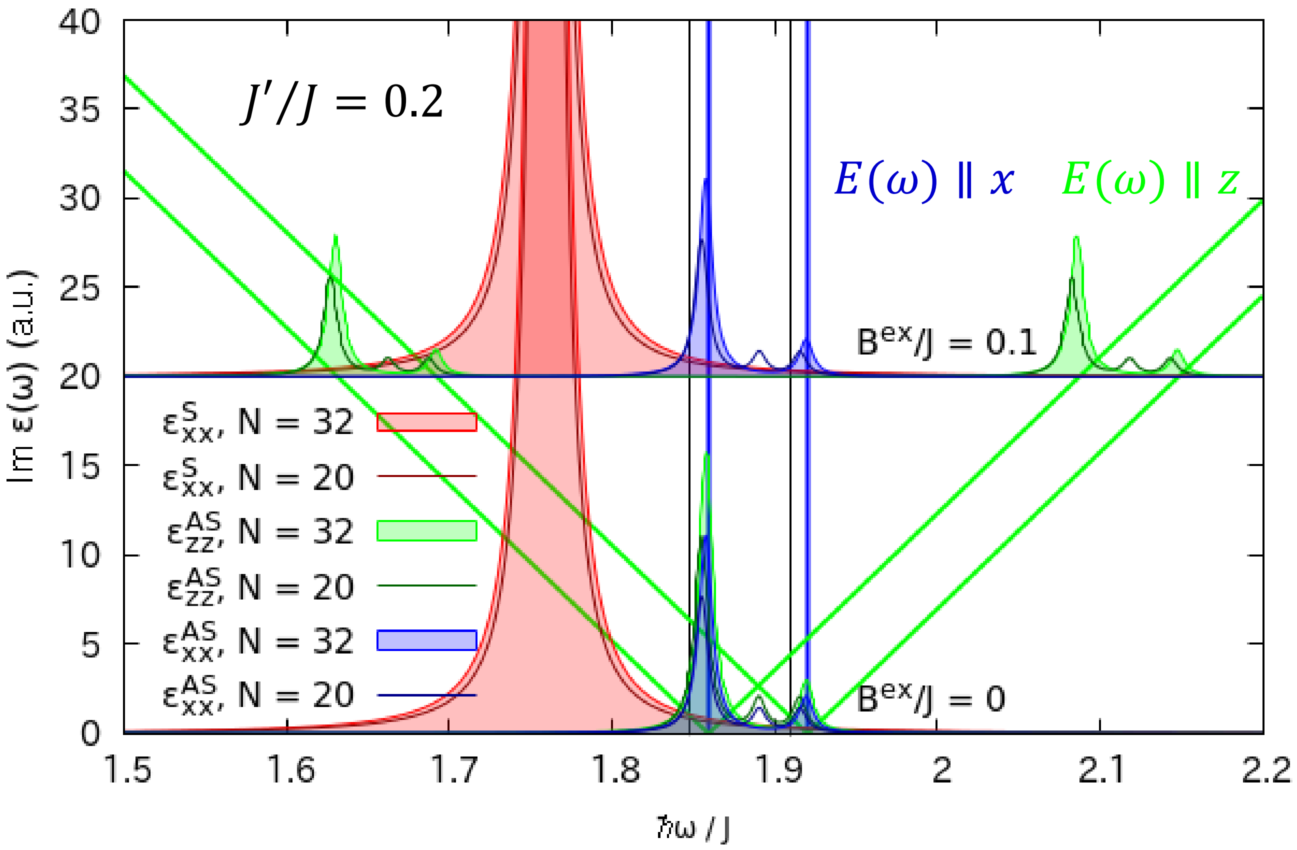
bound sate of two triplets



$S^{\mathrm{T}} = 0$ 4 states (one of them $E(\omega) \perp z$ active)

K. Totsuka *et al*, Phys. Rev. Lett. **86** 520 (2001)

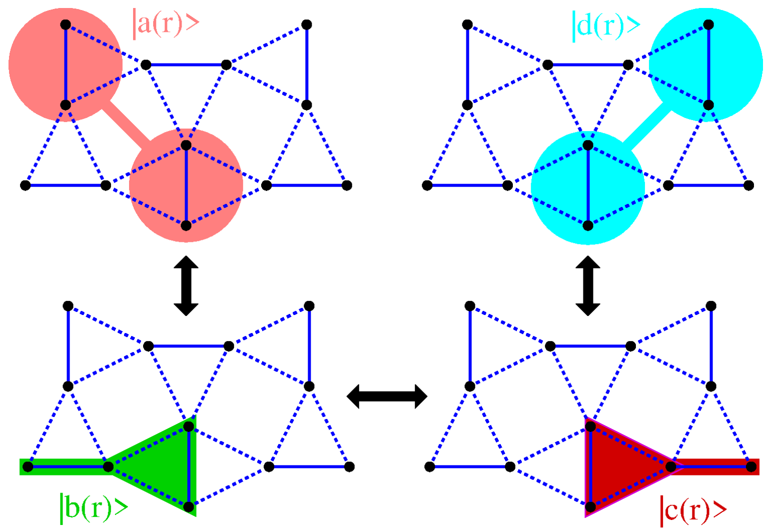
Comparison with the perturbation calculation



$S^T = 1$ bound state are electro active.

3rd order of perturbation

bound sate of two triplets



$S^{\mathrm{T}} = 1$ 4 states

two of them $E(\omega) \perp z, E(\omega) \parallel z$ active

K. Totsuka *et al*, Phys. Rev. Lett. **86** 520 (2001)

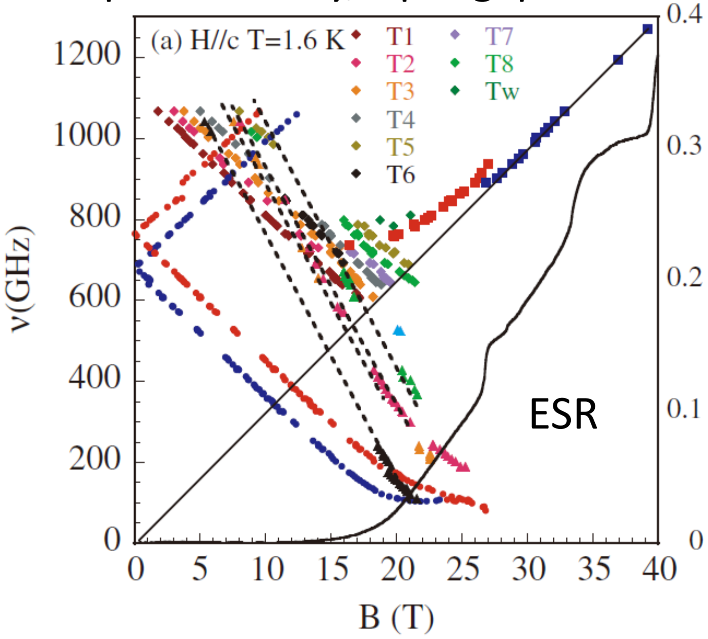
Absorption of spin excitation in $SrCu_2(BO_3)_2$

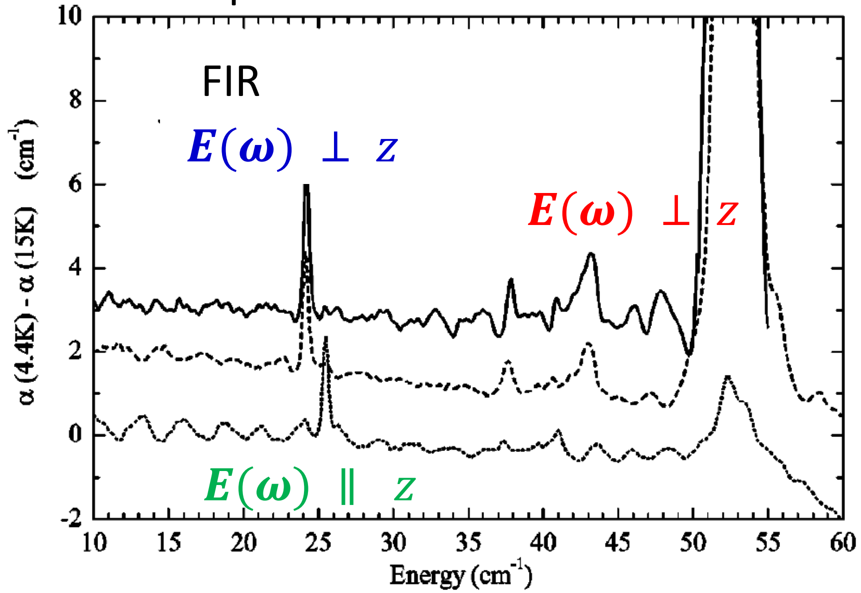
spin gap excitation

transition from singlet g.s. to triplet excitations   forbidden transition in Heisenberg model

c.f. Anisotropic terms induce magneto active mode. *e.g.* M. Oshikawa , JPSJ **72** Suppl. B 36 (2003), T. Saka and H. Shiba JPSJ **63** 867 (1994)

Experimentally, spin gap and bound states of triplet excitations have been observed in absorption





electro-active magnetic excitations

spin gap excitation

$24.2\ \mathrm{cm}^{-1}$ $E(\omega)\ \perp\ z$

$25.7\ \mathrm{cm}^{-1}$ $E(\omega)\ \parallel\ z$

bound states of two triplets

$37.5, 43.0, 52.3, 53.5\ \mathrm{cm}^{-1}$

$E(\omega)\ \perp\ z$

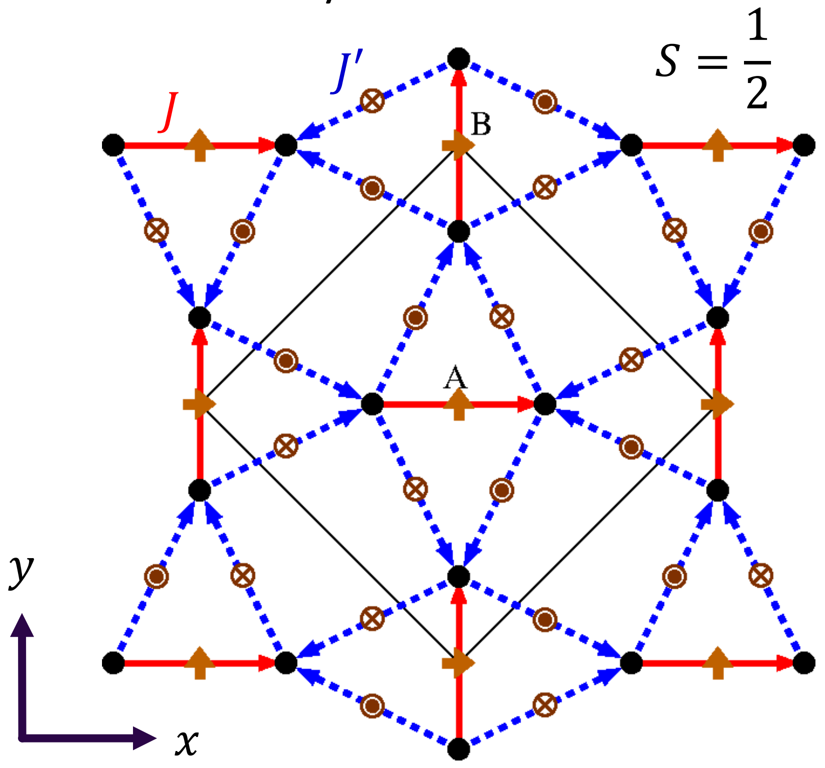
H. Nojiri *et al*, J. Phys. Soc. Jpn. **68** 2906 (1999) , ibid. **72** 3243 (2003)

T. Rõõm *et al*, Phys. Rev. B **61** 14342 (2000), ibid **70** 14417 (2004) , O. Cepas *et al*, Phys. Rev. Lett. **87** 167205 (2001)

Purpose : clarify the origin of the spin gap excitation (selection rule of absorption)

Effects of Dzyaloshinskii-Moriya interaction

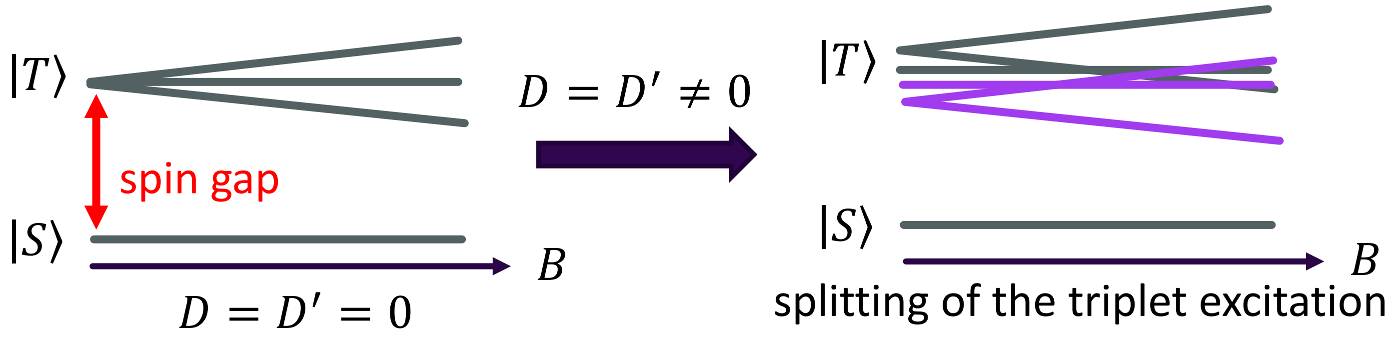
Shastry-Sutherland model



$$\hat{H} = J \sum_{\text{n.n.}} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + J' \sum_{\text{n.n.n.}} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

$$\hat{H}_{\text{DM}} = \sum_{\text{n.n.}} \boldsymbol{D}_{ij} \cdot (\boldsymbol{S}_i \times \boldsymbol{S}_j) + \sum_{\text{n.n.n.}} \boldsymbol{D'}_{ij} \cdot (\boldsymbol{S}_i \times \boldsymbol{S}_j)$$

$$\boldsymbol{D}_{ij} = \begin{cases} (0, D, 0) & \text{A bond} \\ (D, 0, 0) & \text{B bond} \end{cases} \qquad \boldsymbol{D'}_{ij} = (0, 0, \pm D')$$



O. Cepas et al, Phys. Rev. Lett. **87** 167205 (2001), K. Kodama et al, J. Phys.: Condens. Matter **17** L61 (2005)

Electro active magnetic excitation in $SrCu_2(BO_3)_2$

The effects of DM interactions

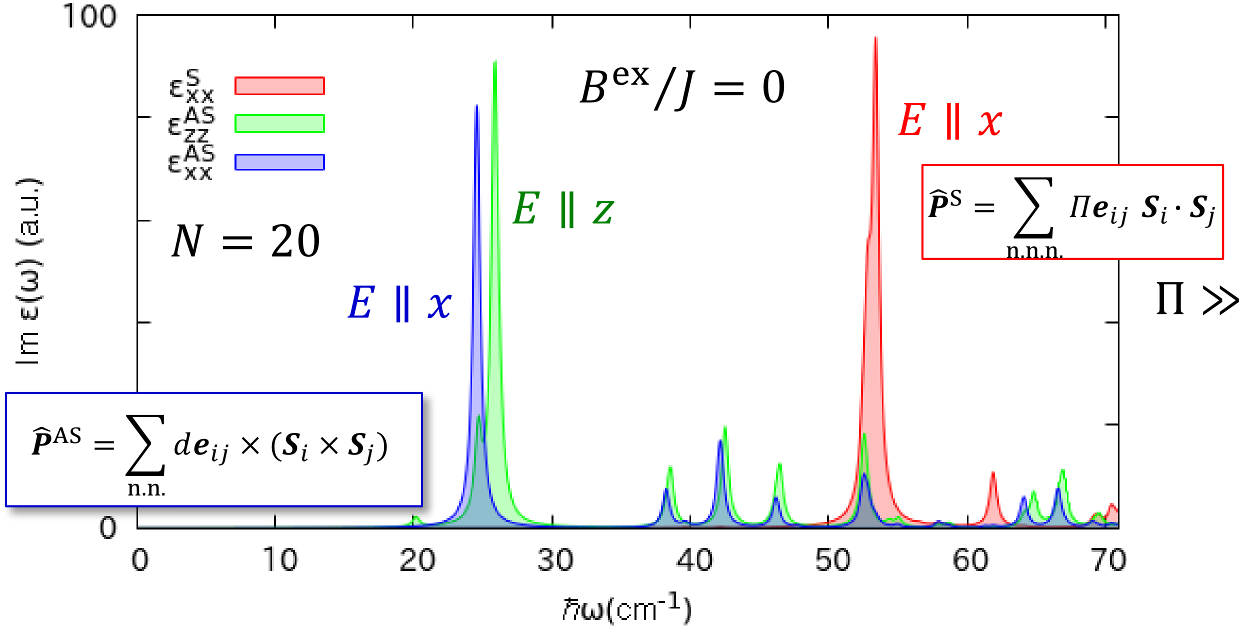
$$\hat{H}_{\mathrm{DM}} = \sum_{\mathrm{n.n.}} \boldsymbol{D}_{ij} \cdot (\boldsymbol{S}_i \times \boldsymbol{S}_j) + \sum_{\mathrm{n.n.n.}} \boldsymbol{D'}_{ij} \cdot (\boldsymbol{S}_i \times \boldsymbol{S}_j)$$

$$\boldsymbol{D}_{ij} = \begin{cases} (0, D, 0) & \text{A bond} \\ (D, 0, 0) & \text{B bond} \end{cases}$$

$$\boldsymbol{D'}_{ij} = (0, 0, \pm D')$$

$$J = 85\mathrm{K} \sim 59\ \mathrm{cm}^{-1}, \frac{J'}{J} = 0.635, \frac{D}{J} = 0.034, \frac{D'}{J} = 0.02,$$

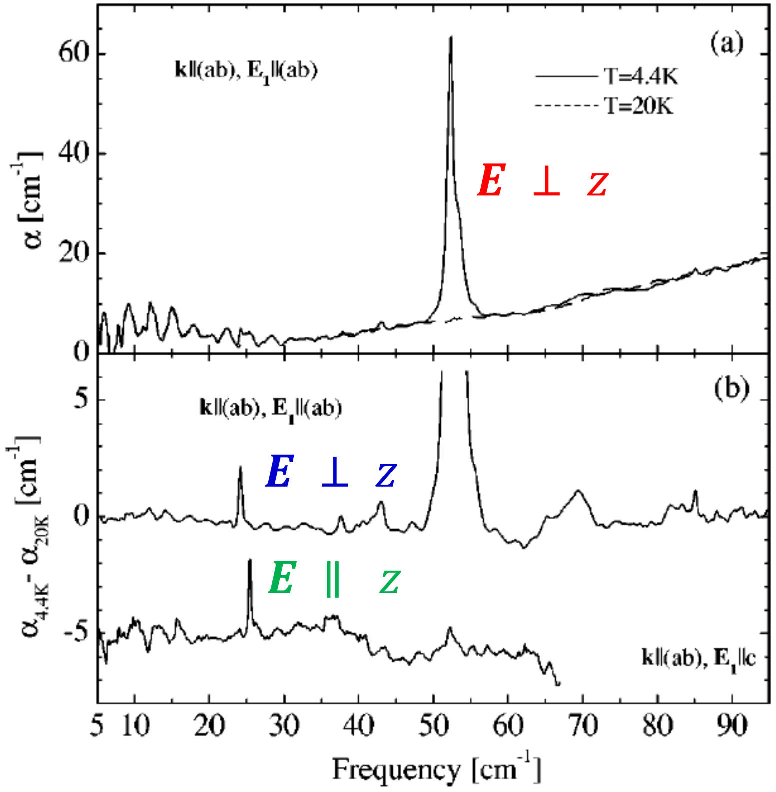
K. Kodama *et al*, J. Phys.: Condens. Matter **17** L61 (2005)



$\Pi \gg d$

FIR

T. Rõõm *et al*, PRB **61** 14342 (2000), ibid **70** 14417 (2004)



The peak positions and selection rules of the main peaks are consistent with FIR experiment.

The magneto active excitations

DM interactions allow magneto active excitation

T. Sakai *et al.* J. Phys. Soc. Jpn. **69** 3521 (2000)

dynamical magnetic susceptibilities

$$\mu_{\alpha\alpha}(\omega) \propto \left\langle 0 \left| \widehat{M}_\alpha^\dagger \frac{1}{\omega + E_0 + i\varepsilon - \widehat{H}} \widehat{M}_\alpha \right| 0 \right\rangle$$

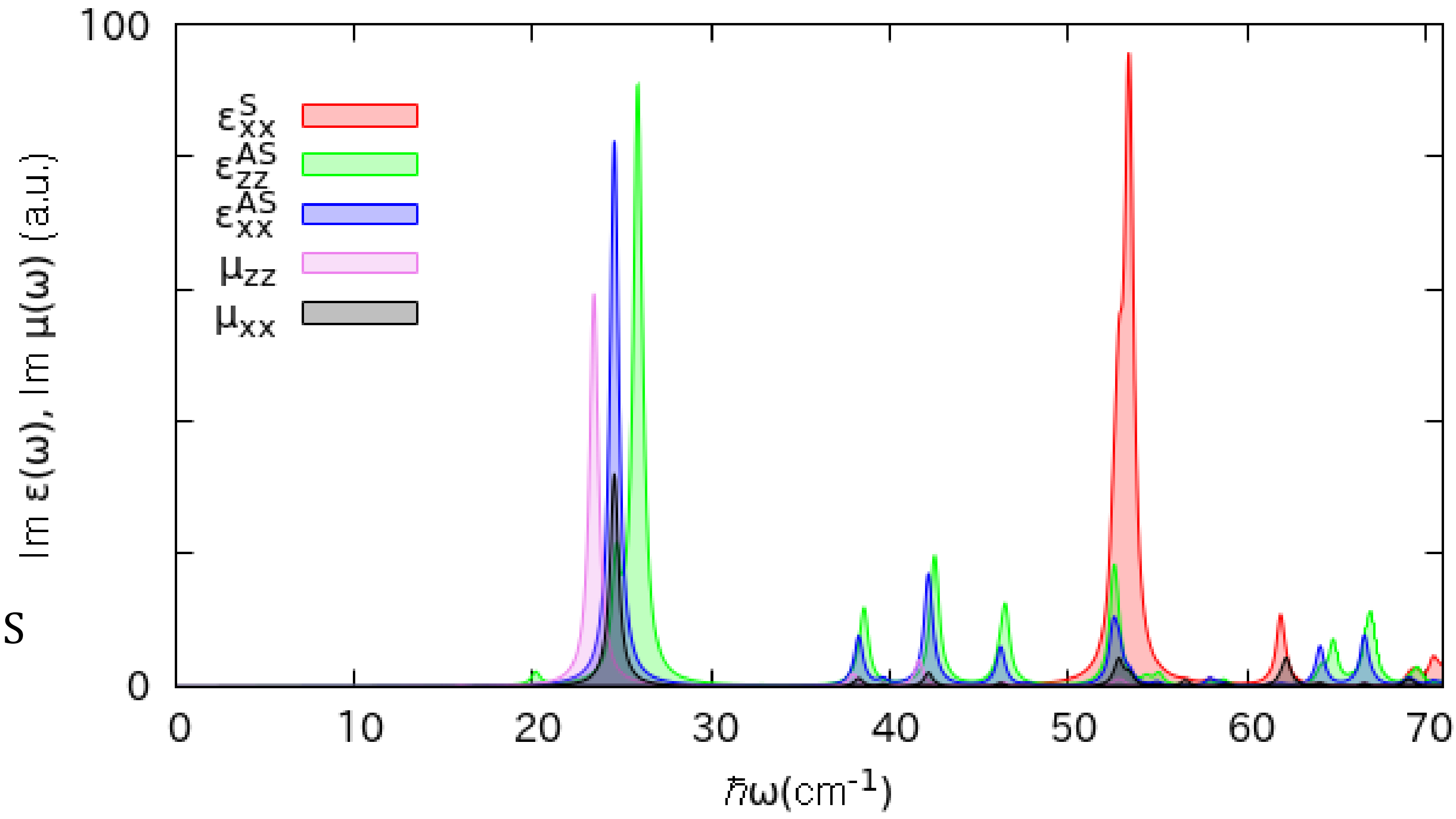
$$\widehat{\boldsymbol{M}} = \sum \boldsymbol{S}_i \qquad \alpha = x, y, z$$

dynamical electric susceptibility

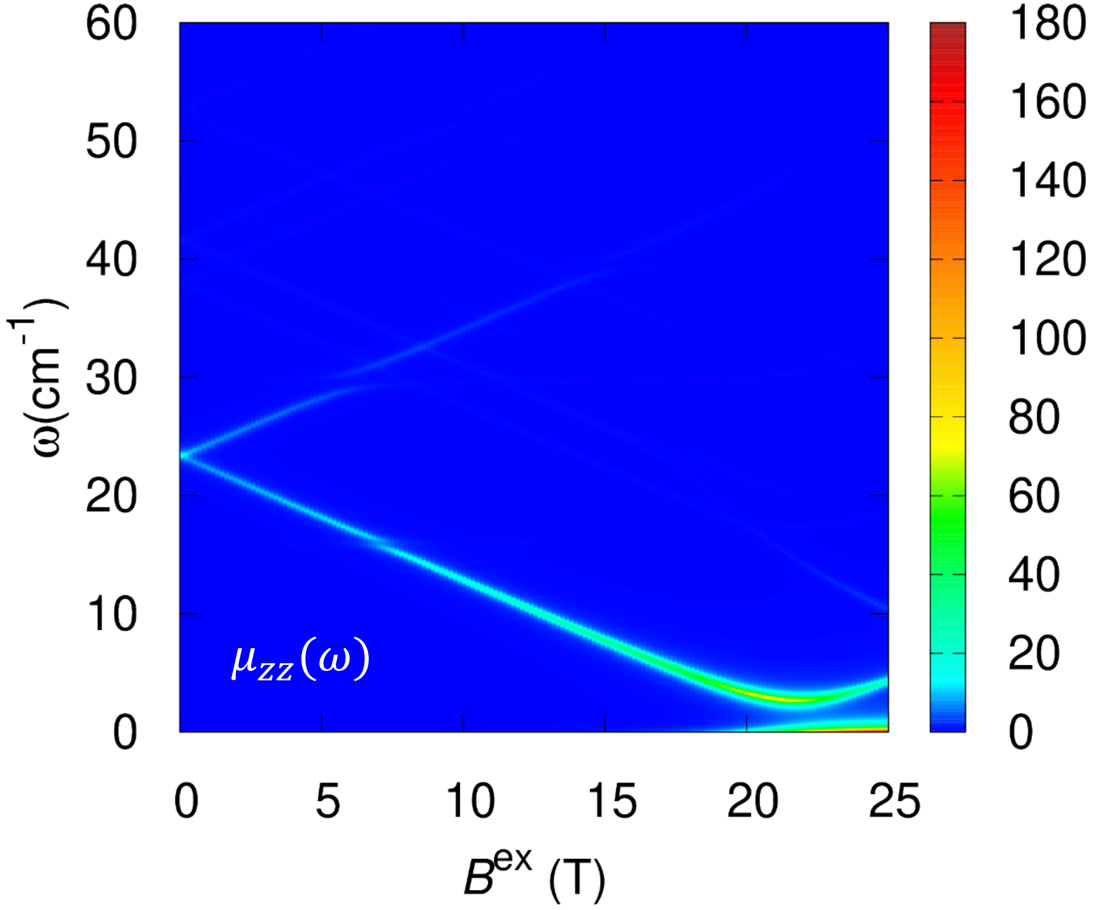
$$\varepsilon_{\alpha\alpha}^{\beta}(\omega) \propto \left\langle 0 \left| \widehat{P}_\alpha^{\beta\dagger} \frac{1}{\omega + E_0 + i\varepsilon - \widehat{H}} \widehat{P}_\alpha^{\beta} \right| 0 \right\rangle$$

$$\alpha = x, y, z \qquad \beta = \mathrm{AS}, \mathrm{S}$$

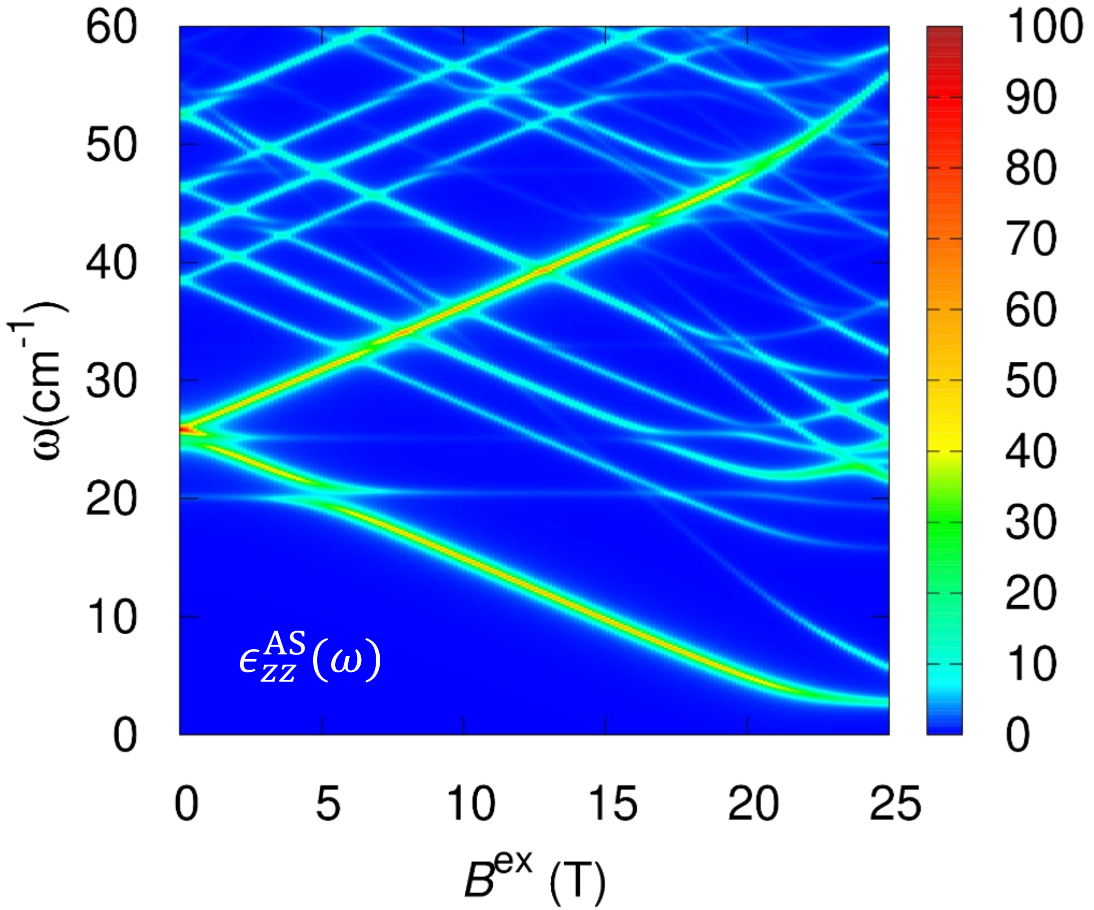
$$J = 85\mathrm{K} \sim 59\ \mathrm{cm}^{-1}, \frac{J'}{J} = 0.635, \frac{D}{J} = 0.034, \frac{D'}{J} = 0.02,$$



Magnetic field dependence $B^{\text{ex}} \parallel z$



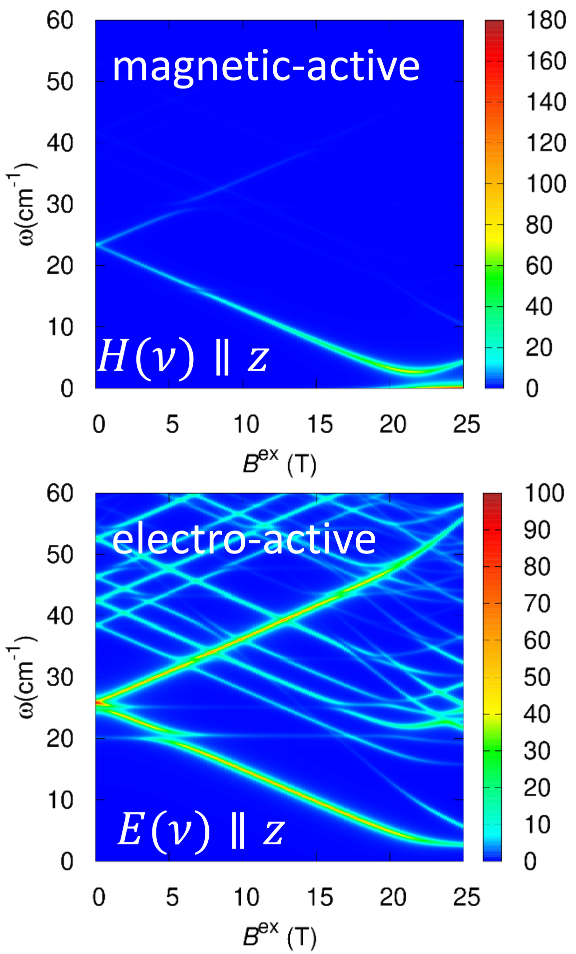
singlet-triplet excitation allowed due DM interaction

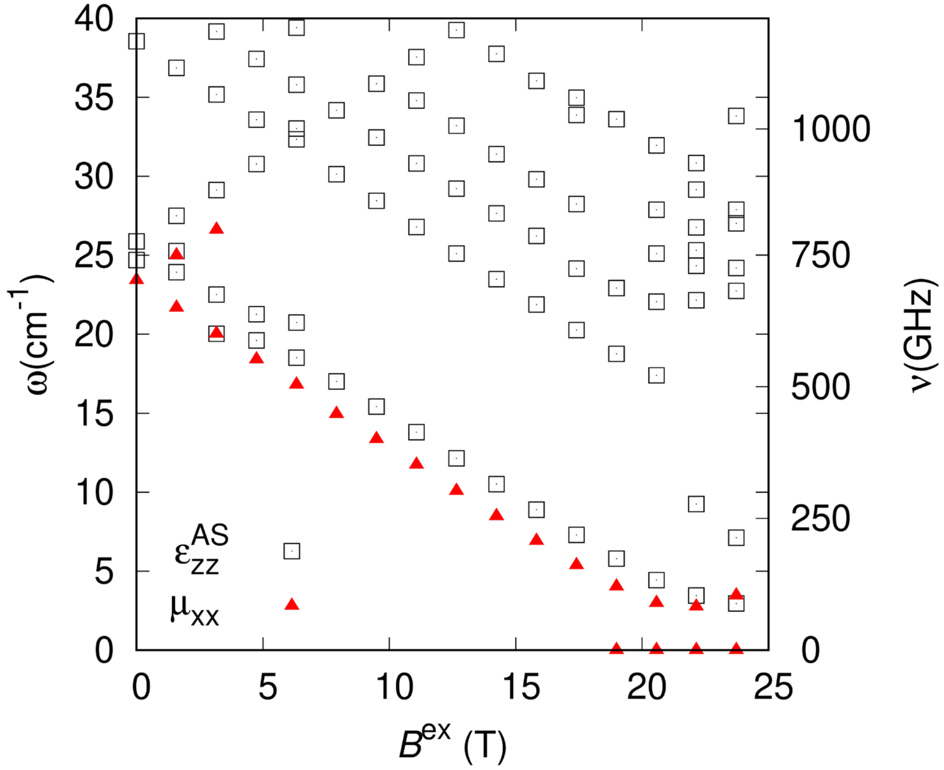


singlet-triplet excitation induced by electric components of light

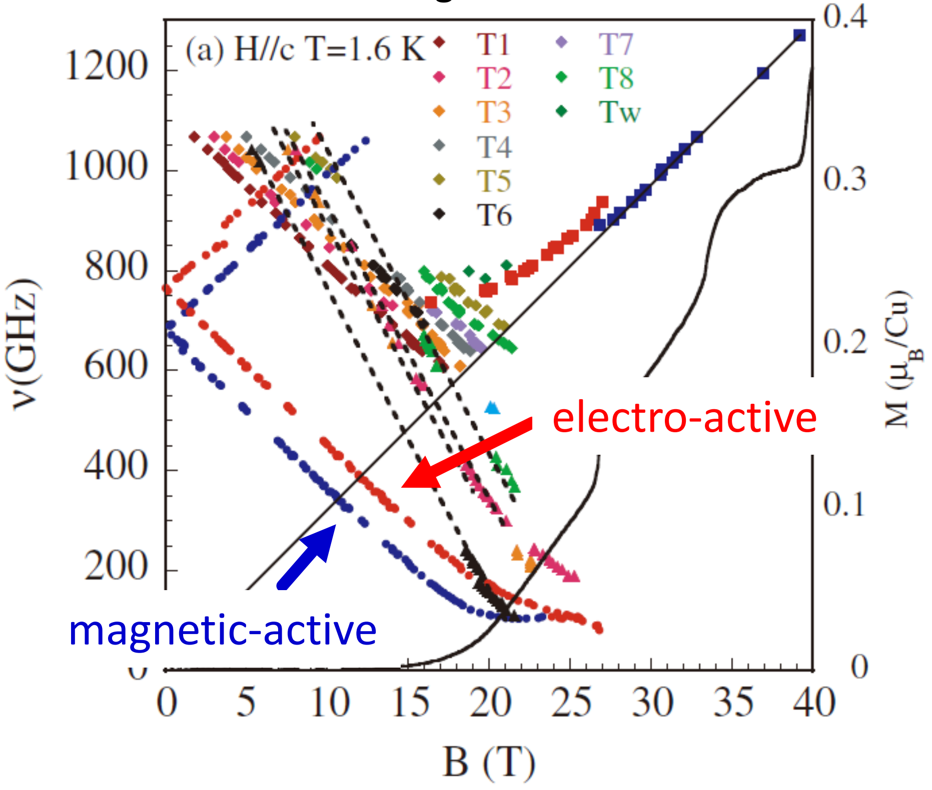
bound states of two triples induced by electric components of light

Comparison with ESR





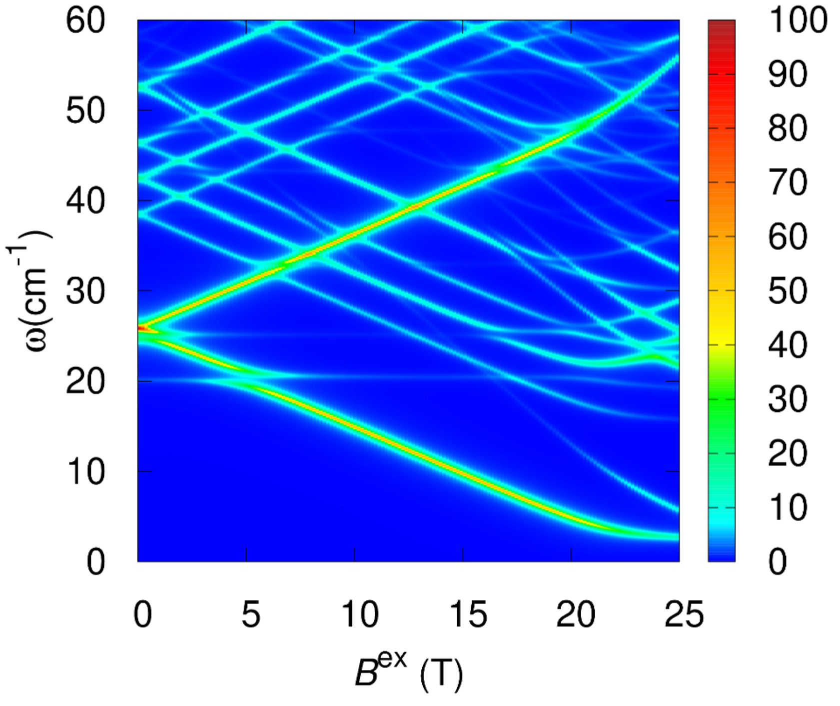
ESR
external magnetic field $B \parallel z$



H. Nojiri *et al*, J. Phys. Soc. Jpn. **72** 3243 (2003)

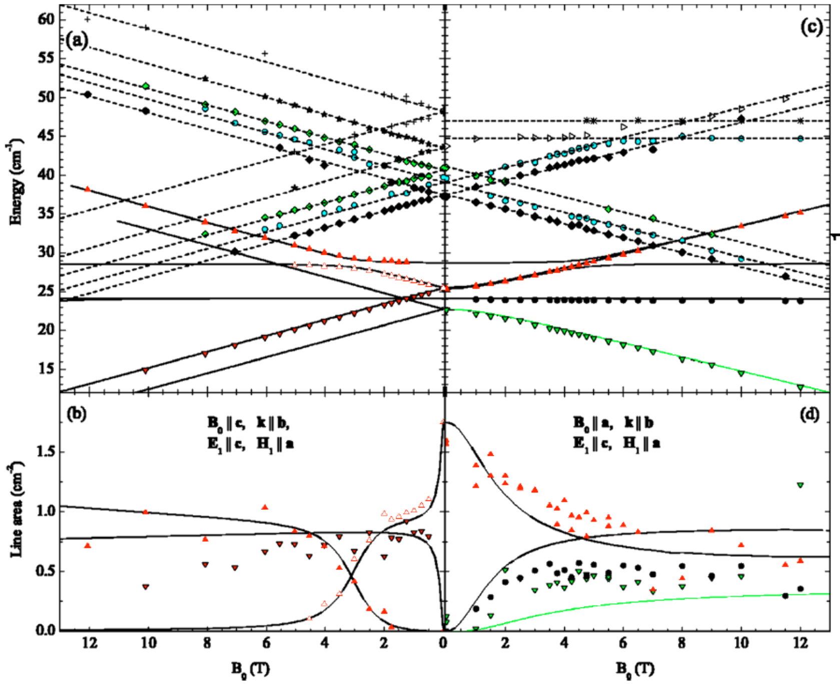
Comparison with FIR

$\epsilon_{zz}^{\mathrm{AS}}(\omega)$



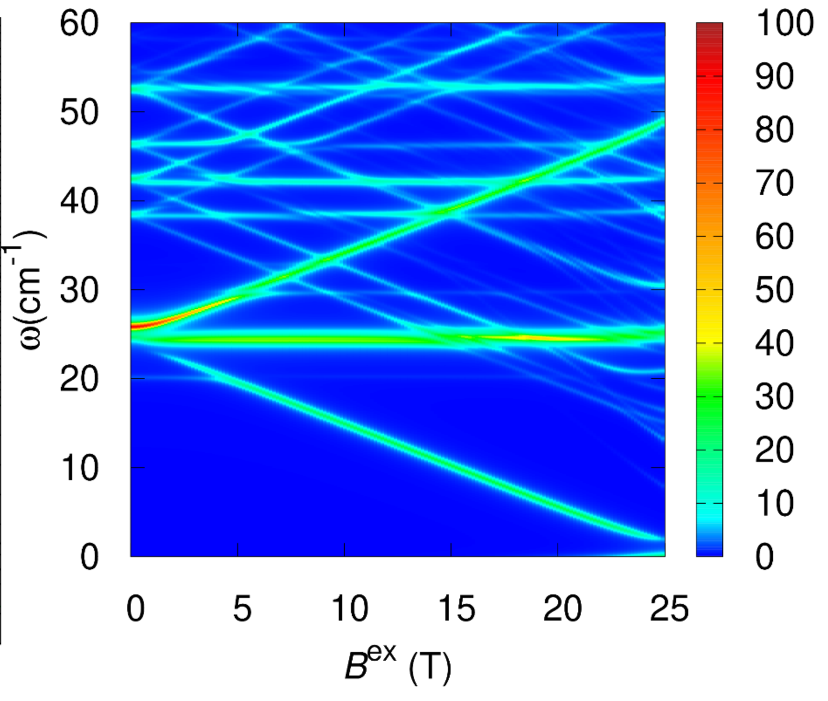
$B^{\mathrm{ex}} \parallel z$

linearly polarized light



T. Rõõm *et al*, PRB **70** 14417 (2004)

$\epsilon_{zz}^{\mathrm{AS}}(\omega)$



$B^{\mathrm{ex}} \parallel x$

Conclusion

Conclusion

- The spin-electron coupling and DM interactions allow an absorption in $SrCu_2(BO_3)_2$.
  - The selection rule is consistent with experimental observation
  - Magnetic excitations are allowed by the spin-electron coupling even in Heisenberg model.

c.f. $TlCuCl_3$: S. Kimura *et al.* J. Magn. Magn. Mater. **310** 1218 (2007), $KCuCl_3$: S. Kimura *et al.* Appl. Magn Reson. **46** 1035 (2015)

The clarification of magnetic excitation processes induced by $E(\omega)$ can play an important role in the investigation of magnetic excitation in non-magnetic ground states.

Open issues

- The selection rule in the plaquette singlet state

  pressure induced phase transition in in $SrCu_2(BO_3)_2$ T. Sakurai *et al*, J. Phys. Soc. Jpn. **87** 033701 (2018)
- The proposal of the novel magnetic excitations and magnetoelectric effects in spin systems