1. Introduction

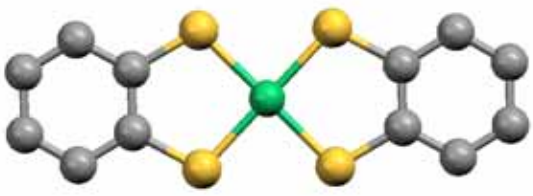
Molecular magnets

- Anisotropic interaction
  ⇨ Low dimensionality, frustrated system
- Designability of molecules
  ⇨ Controllability of magnetic interactions

We used the following two transition metal complexes

$M(9S3)_2^{2+}$
(M = Ni,Co)

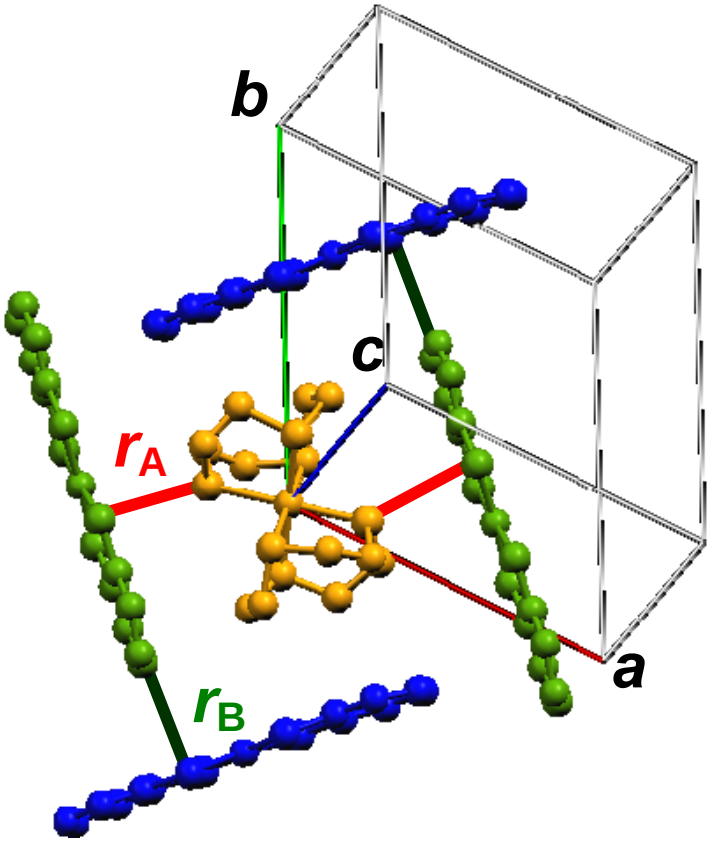
- 9S3 = 1,4,7- trithiacyclononane
- Octahedral coordination complex
- *S* = 1 (M=Ni) or S = 1/2 (M=Co)
- Strong inter-molecular interaction through sulfur atom of 9S3



$Ni(bdt)_2^-$

- bdt = 1,2-benzenedithiolate
- *S* = 1/2
- Anisotropic interaction
  Strong: through sulfur atom
  Weak: through benzene ring

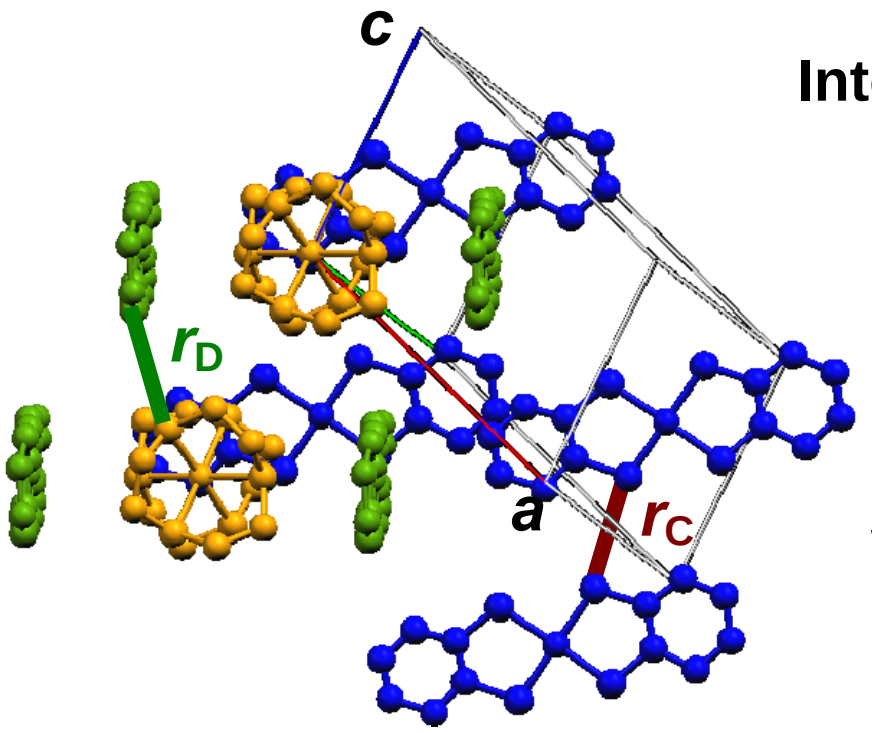
2.Crystal Structure



Cage-like structure
(in (*a*+*c*)-(*a*+*b*) plane)

S-S contact
r_A = 3.77Å
Strong interaction

CH–S contact
r_B = 3.75Å
Weak interaction

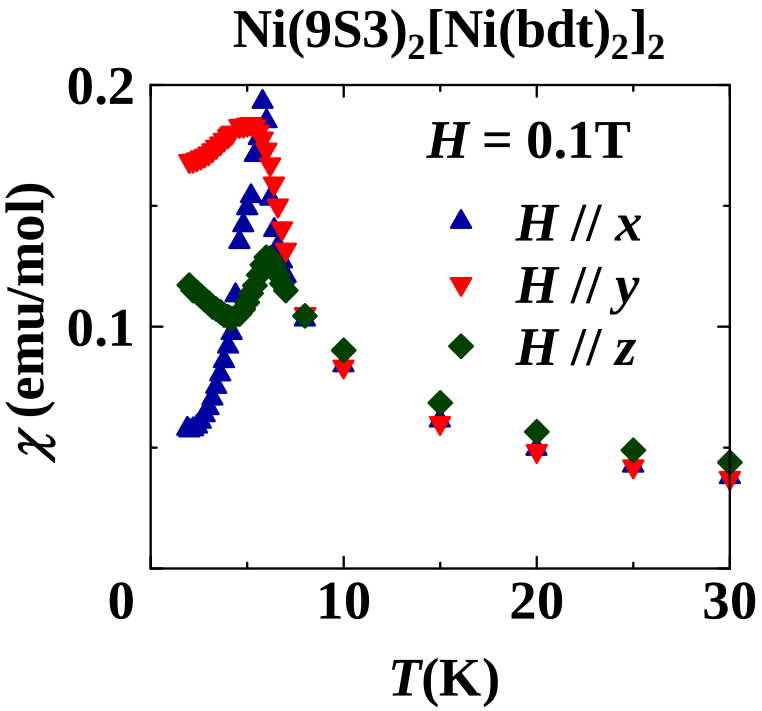


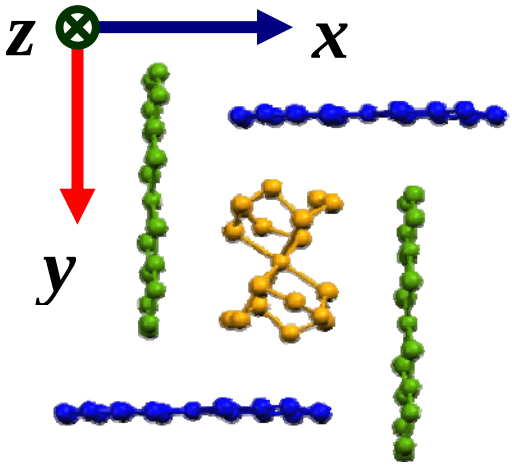
Inter-plane stacking

S-S contact
r_C = 3.78Å
Strong interaction

CH–S contact
r_D = 3.98Å
Weak interaction

3.Magnetic Susceptibility (M=Ni)





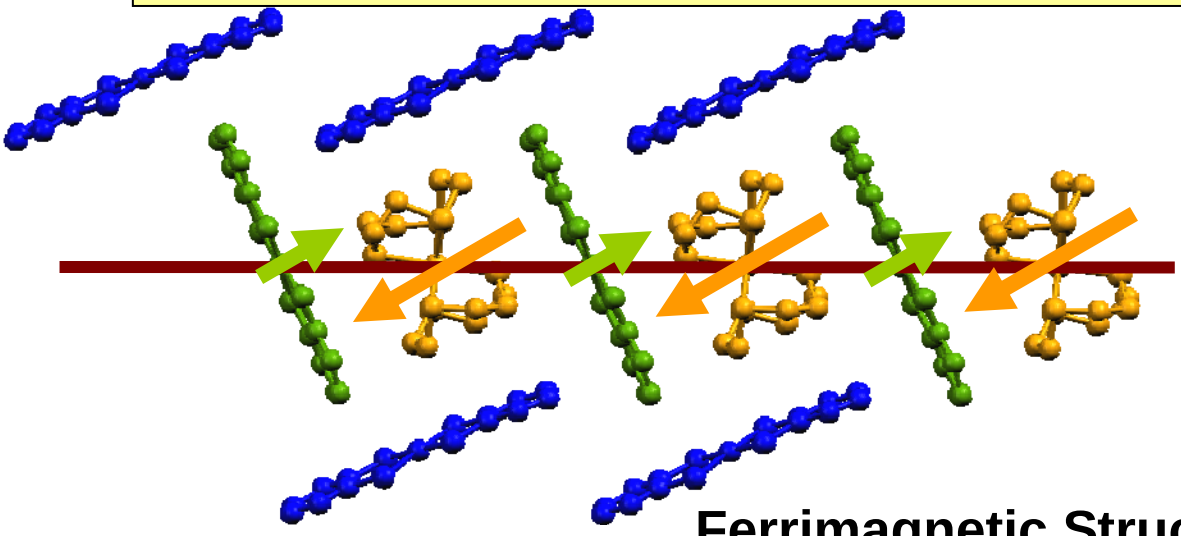
Θ = -6.5 K

T_N = 5.8 K

χ of H // x and y increase at 6K < T <

Caused by intra-chain ordering of

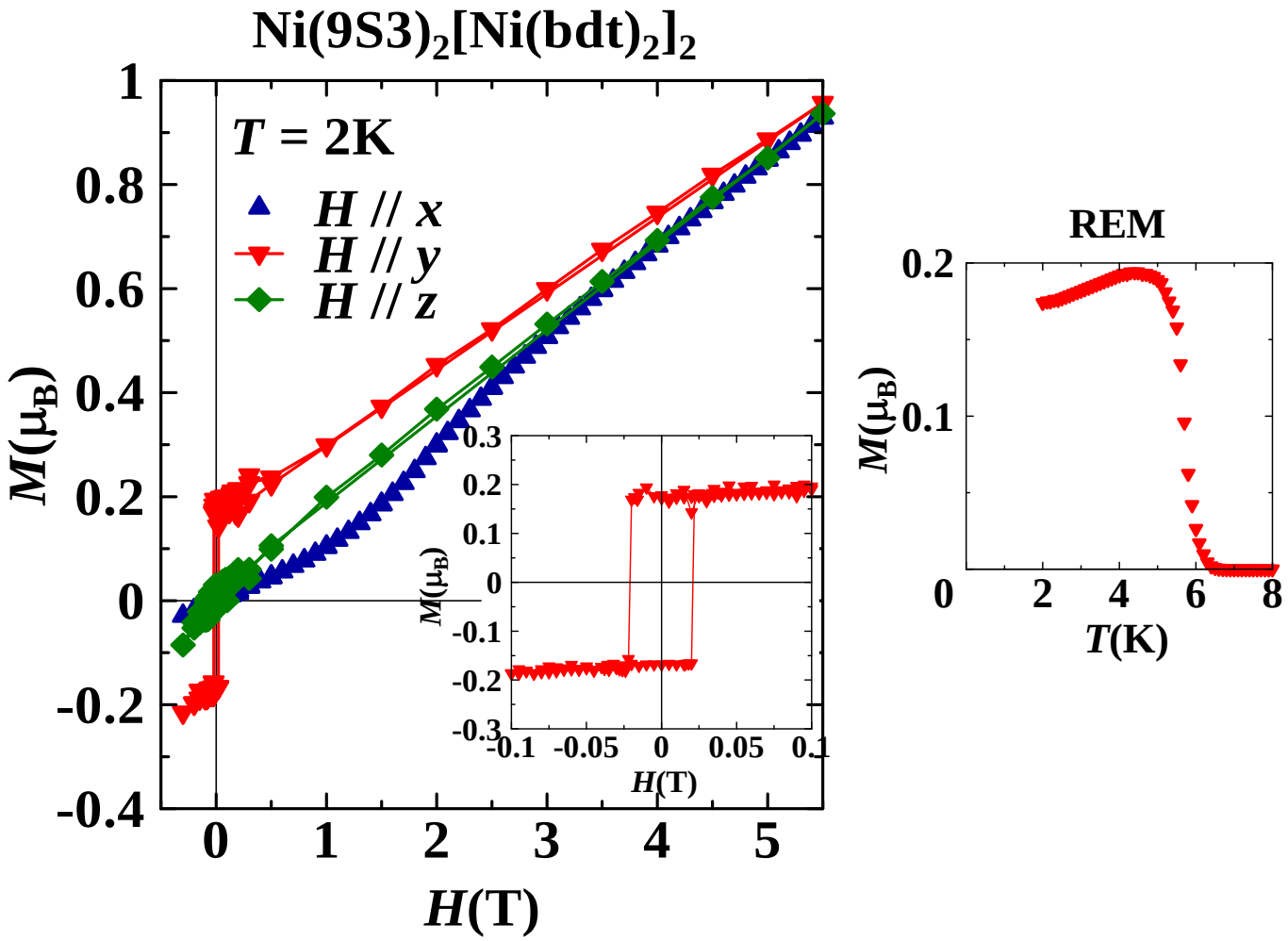
$Ni(9S3)_2^{2+}$-$Ni(bdt)2^{-}$ ferrimagnetic chain



Ferrimagnetic Structure

Below T_N = 5.8K, weak inter-chain interactions causes antiferromagnetic transition.

4.Magnetization Curve (M=Ni)

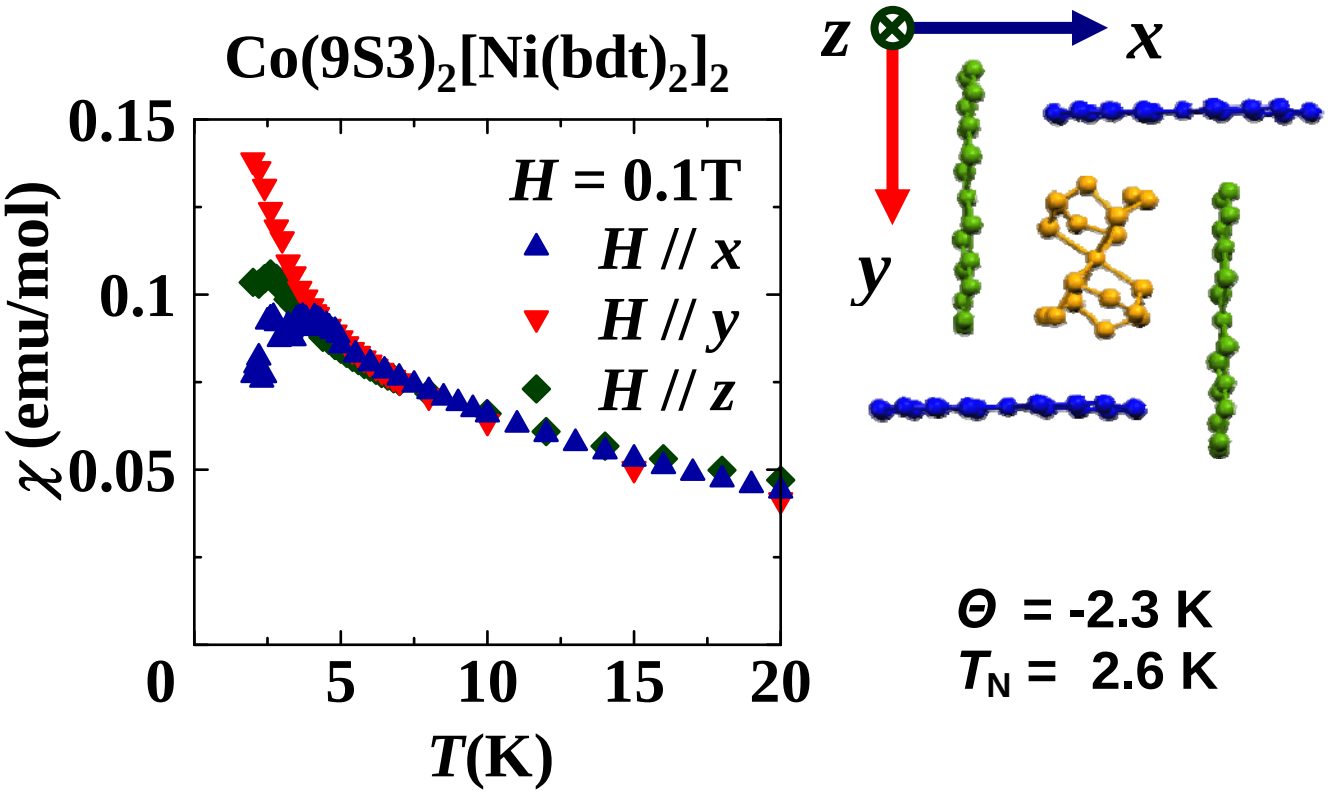


Weak-ferromagnetism (*H // y*)

Remanent magnetization: 0.2 μ_B

Coercive force: 200 Oe

5.Magnetic Susceptibility (M=Co)

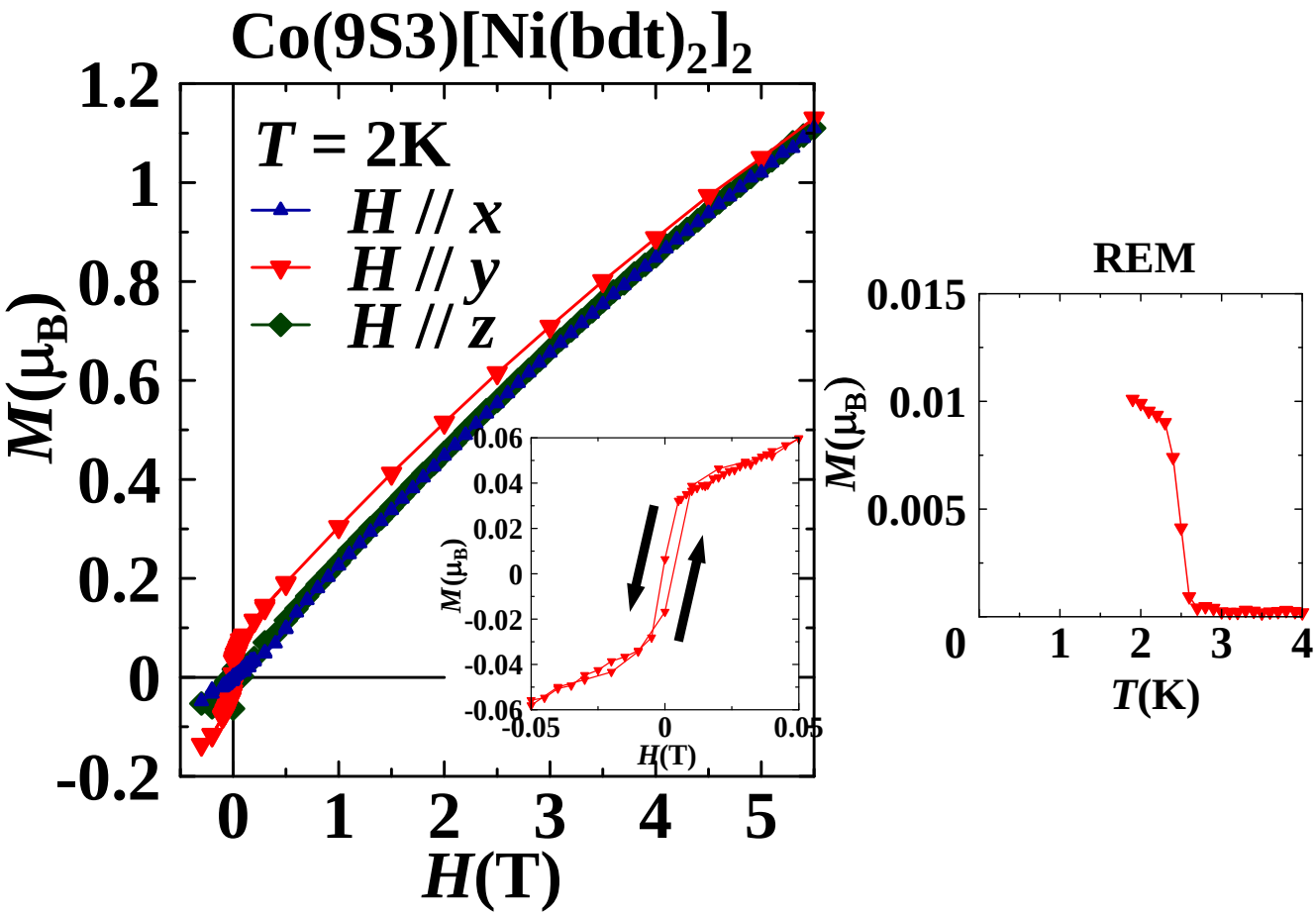


No remarkable increase above T_N was observed.

⇧

In the case of Co, there is no ferrimagnetic structure.

6.Magnetization Curve (M=Co)



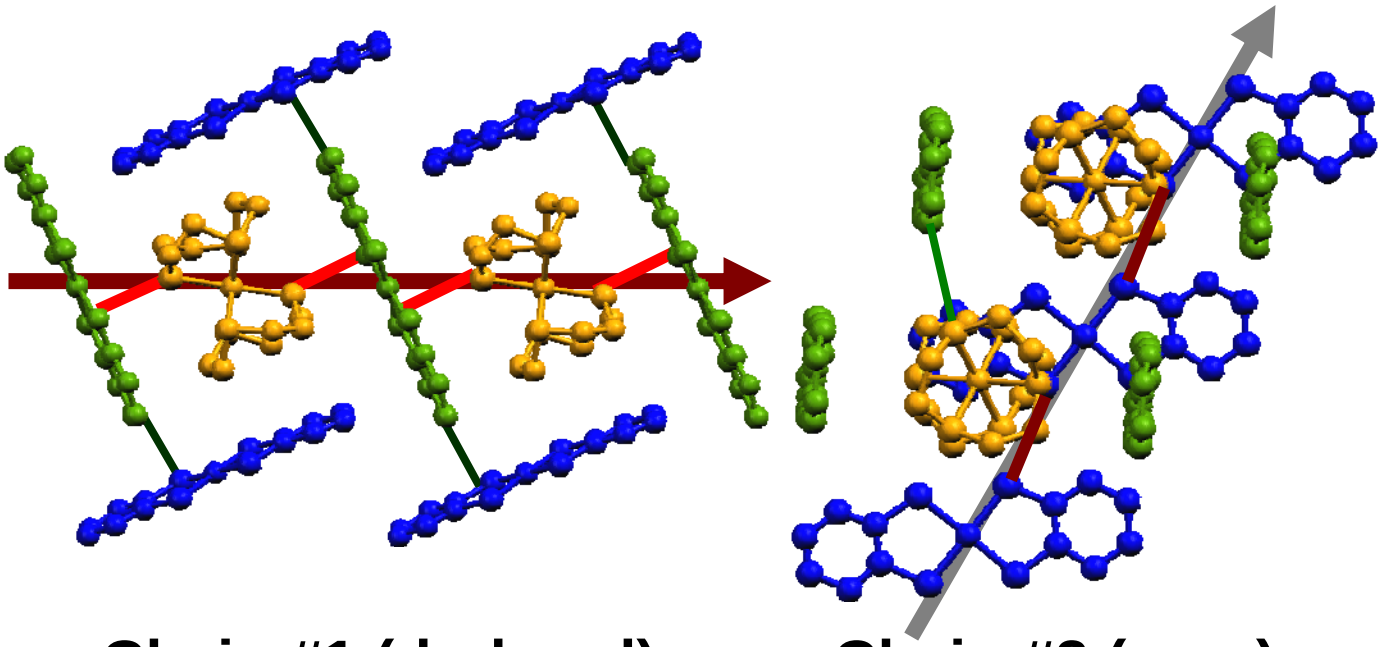
Quite weak weak-ferromagnetism (*H // y*)
Remanent magnetization: 0.01 μ_B
Coercive force: 10 Oe

This weakening of weak-ferromagnetism suggests
Interaction between $M(9S3)_2^{2+}$ and $Ni(bdt)_2^{-}$
or
Ferrimagnetic chain structure
plays important role in weak-ferromagnetism.

7.Origin of Weak-Ferromagnetism

First, we consider about the Ni salt.

The salt consists of two magnetic chains.

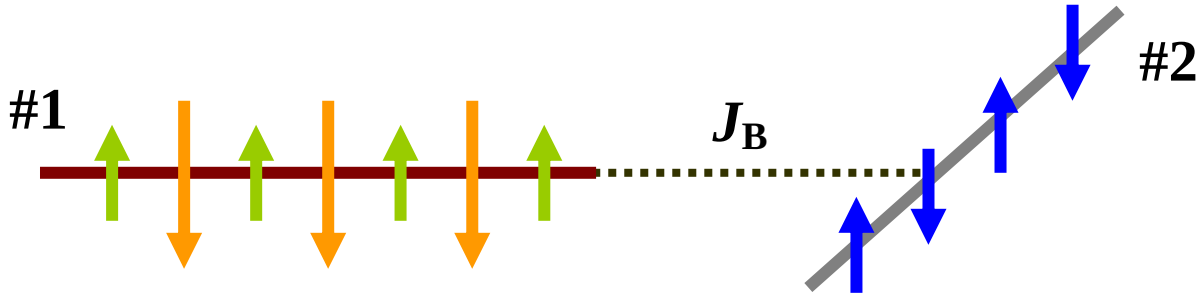


Chain #1 (dark red)　　Chain #2 (gray)

J_A: strong (intra-chain)　　J_C: strong (intra-chain)

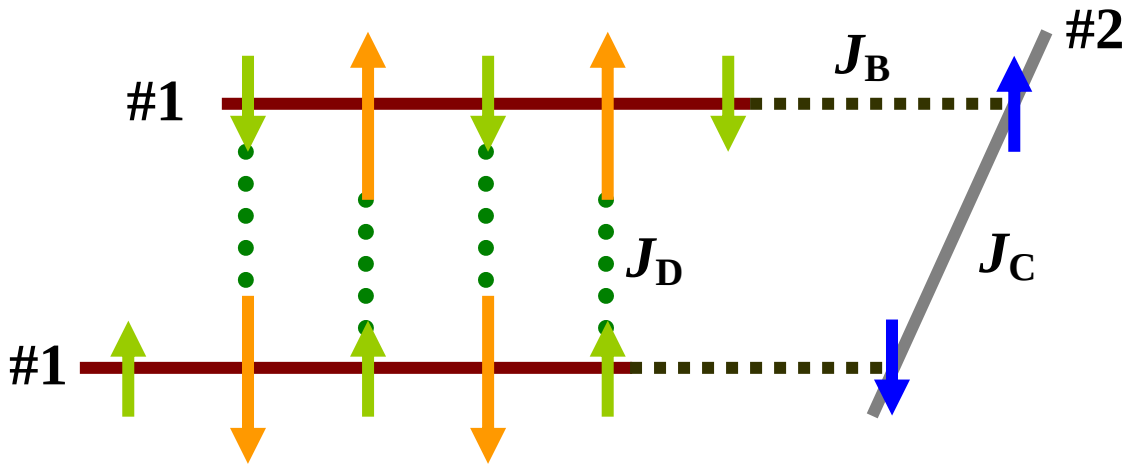
J_B: weak (#1 - #2)　　J_D: weak (#1 - #1)

Judging from the crystal structure, $|J_A|, |J_C| >> |J_C| > |J_D|$



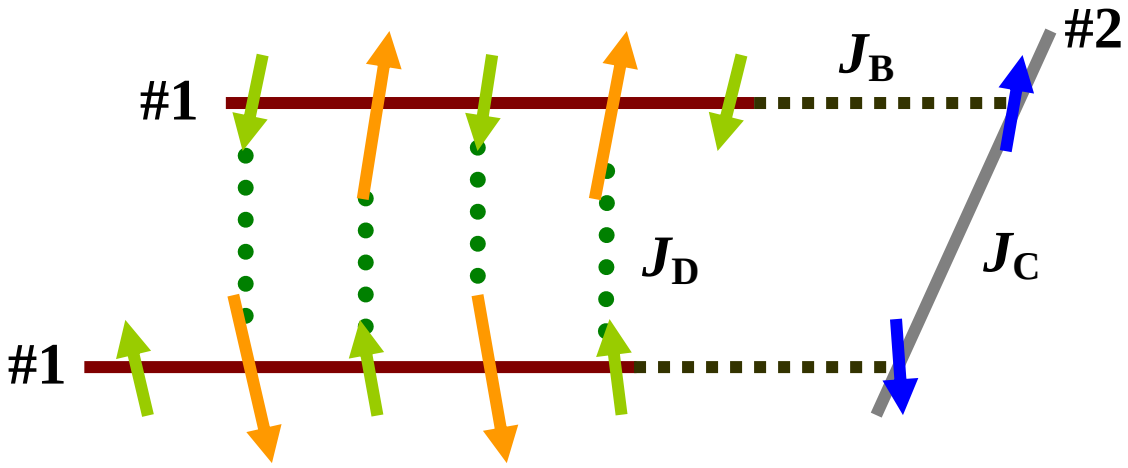
In plane magnetic structure

Considering about inter-plane interactions



Competition of J_B and J_D occurs

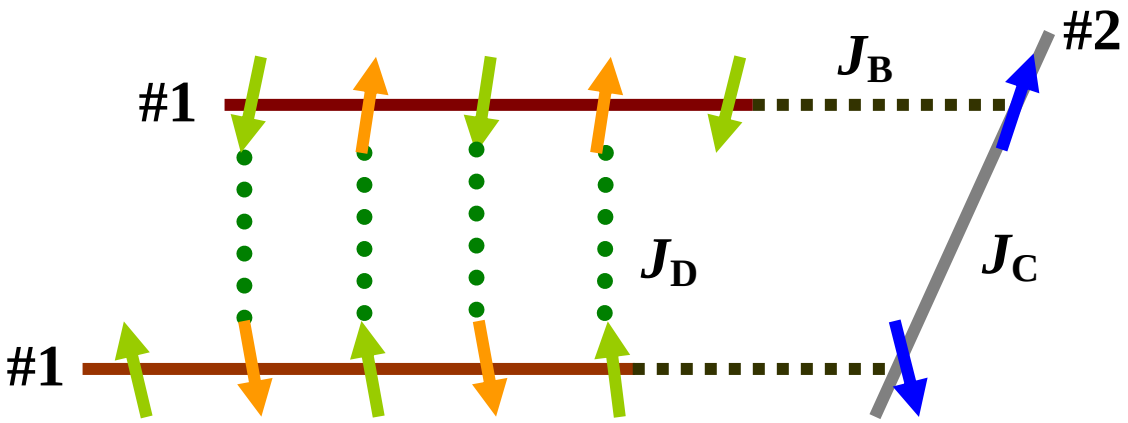
To decrease the competition, spins cant



Weak-ferromagnetism appears

In the case of Co (*S*=1/2) salt

- **ferrimagnetic structure disappears**
  - ⇨ **Chains #1 don't show weak-ferromagnetism**
- **#1 - #1 inter-chain interaction (J_D) weaken**
  - ⇨ **Canting angle decreases**



Only canting of chains #2
contribute weak-ferromagnetism

Quite weak
Weak-Ferromagnetism

Summary

New weak-ferromagnet $M(9S3)_2[Ni(bdt)_2]_2$

M=Ni

Transition temperature T_N = 5.8 K
Weiss temperature Θ = - 6.5K
Coercive foerce H_C = 200 Oe
Remanent Magnetization M_{REM} = 0.2 μ_B

M=Co

Transition temperature T_N = 2.6 K
Weiss temperature Θ = - 2.3K
Coercive foerce H_C = 10 Oe
Remanent Magnetization M_{REM} = 0.01 μ_B

- The origin of weak-ferromagnetism

  Competition of
  two antiferromagnetic interactions

- Weak weak-ferromagnetism of Co salt

  Disappearance of ferrimagnetic structure
  Decreasing of competition