

2022.06.15 Lecture 10

Lecture on

10:25 – 11:55

Magnetic Properties of Materials

磁性 (Magnetism)

Institute for Solid State Physics, University of Tokyo

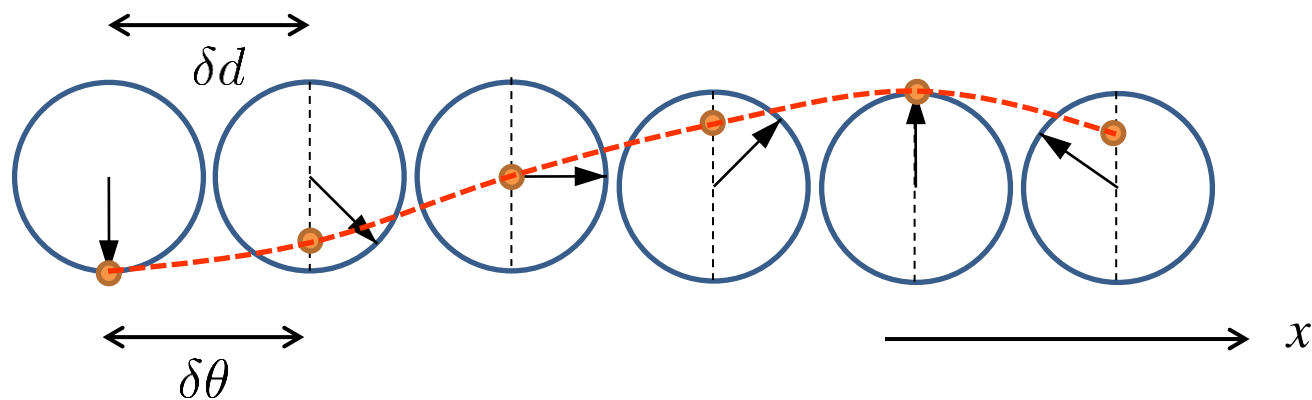
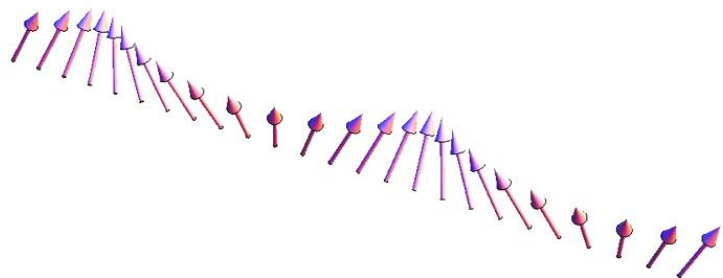
Shingo Katsumoto

- Anti-ferromagnetic Heisenberg model : parallel field susceptibility
- Spin flop and metamagnetic transition
- Ferrimagnetism
- Molecular-field approximation
- Helimagnetism
- Spin wave

- Spin wave (classical) in a ferromagnet
- Quantization of spin wave (magnon)
- Magnons in an anti-ferromagnet
- Magnon approximation for weak excitations in ferro- and anti-ferromagnets
- Nambu-Goldstone mode in phase transition
- Experiments on magnons

Spin wave from phase shift of spin precessions

<https://www.youtube.com/watch?v=pWQ3r-2Xjeo>



snapshot $S_x = S_0 \cos(n\delta\theta) = S_0 \cos\left(x \frac{\delta\theta}{\delta d}\right) = S_0 \cos kx$

$$S = S_x + iS_y = S_0 \exp(ikx)$$

cf. Bloch electrons → Magnons can be described in magnetic Brillouin zone.

Total spin $S = \sum_i S_i$ Heisenberg equation: $i\hbar \frac{\partial S}{\partial t} = [S, \mathcal{H}]$ Such a motion of macroscopic magnetic moment can be confirmed by Ferromagnetic resonance (FMR)

Phase shifts of precessions with sites:

$S_{ix} = A \cos(\omega_0 t + \theta_i), \quad S_{iy} = A \sin(\omega_0 t + \theta_i)$ Then a snapshot should be expressed in a Fourier form.

However for ω_0 we need to consider the spin-spin interaction.

Equations of motion in the momentum space

Fourier transform, inverse Fourier transform:

$$S_{\mathbf{q}x} = \frac{1}{\sqrt{N}} \sum_j S_{jz} \exp(-i\mathbf{q} \cdot \mathbf{r}_j), \quad S_{jz} = \frac{1}{\sqrt{N}} \sum_{\mathbf{q}} S_{\mathbf{q}x} \exp(i\mathbf{q} \cdot \mathbf{r}_j)$$

Heisenberg Hamiltonian, equation of motion:

$$\mathcal{H} = -2J \sum_{\langle i,j \rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j, \quad i\hbar \frac{\partial \mathbf{S}}{\partial t} = [\mathbf{S}, \mathcal{H}]$$

Substituting the above Fourier transforms into equation of motion, we obtain a set of equations of motion in the momentum space as:

$$\left\{ \begin{aligned} i\hbar \frac{\partial S_{\mathbf{q}x}}{\partial t} &= \frac{4i}{\sqrt{N}} J \sum_{\langle i,j \rangle} S_{iy} S_{jz} \exp(-i\mathbf{q} \cdot \mathbf{r}_i) \{1 - \exp[i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)]\} \\ i\hbar \frac{\partial S_{\mathbf{q}y}}{\partial t} &= -\frac{4i}{\sqrt{N}} J \sum_{\langle i,j \rangle} S_{ix} S_{jz} \exp(-i\mathbf{q} \cdot \mathbf{r}_i) \{1 - \exp[i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)]\}. \end{aligned} \right.$$

Fourier transform of interaction J :

$$J_{\mathbf{q}} = \sum_j J \exp[i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)] \quad (i \text{ can be taken somewhere})$$

Nearest neighbor approximation:

Small angle approximation, i.e.,
replace S_{jz} with S :

$$\left\{ \begin{aligned} \hbar \frac{\partial S_{\mathbf{q}x}}{\partial t} &= 2[J_0 - J_{\mathbf{q}}] S S_{\mathbf{q}y}, \\ \hbar \frac{\partial S_{\mathbf{q}y}}{\partial t} &= -2[J_0 - J_{\mathbf{q}}] S S_{\mathbf{q}x}. \end{aligned} \right.$$

Spin wave (ferromagnetic)

These are the equation we obtained in the last lecture but B :

$$\hbar \frac{d\mathbf{S}_{\mathbf{q}}}{dt} = -[2(J_0 - J_{\mathbf{q}})S + \mu B] \mathbf{e}_z \times \mathbf{S}_{\mathbf{q}}$$

These equation represents precession around z-axis (in Fourier space)

$$\left\{ \begin{array}{l} \hbar \frac{dS_{qx}}{dt} = [2(J_0 - J_{\mathbf{q}})S + \mu B] S_{qy}, \\ \hbar \frac{dS_{qy}}{dt} = -[2(J_0 - J_{\mathbf{q}})S + \mu B] S_{qx}, \\ \hbar \frac{dS_{qz}}{dt} = 0 \end{array} \right.$$

Hence we write

$$S_{qx} + iS_{qy} \propto \exp[-i\epsilon_{\mathbf{q}}t/\hbar]$$

to obtain the excitation energy

$$\underline{\epsilon_{\mathbf{q}} = 2(J_0 - J_{\mathbf{q}})S + \mu B}$$

Remember the message:

$$\left[\begin{array}{l} S_{ix} = A \cos(\omega_0 t + \theta_i), \quad S_{iy} = A \sin(\omega_0 t + \theta_i) \\ \text{However for } \omega_0 \text{ we need to consider the spin-spin interaction.} \end{array} \right]$$

Holstein-Primakoff transformation

Let us consider the **quantization of the spin wave**.

Spin operator: $\mathbf{S} \quad |m\rangle$: eigenfunction of S_z with eigenvalue of m .

We define up/down operator as: $S_{\pm} = S_x \pm S_y$

Then from the properties of spin operator:

$$\left. \begin{aligned} S_+ |m\rangle &= \sqrt{S(S+1) - m(m+1)} |m+1\rangle \\ S_- |m\rangle &= \sqrt{S(S+1) - m(m-1)} |m-1\rangle \end{aligned} \right\} \begin{array}{l} \text{vacuum} \\ |S\rangle \rightarrow (|0\rangle) \end{array}$$

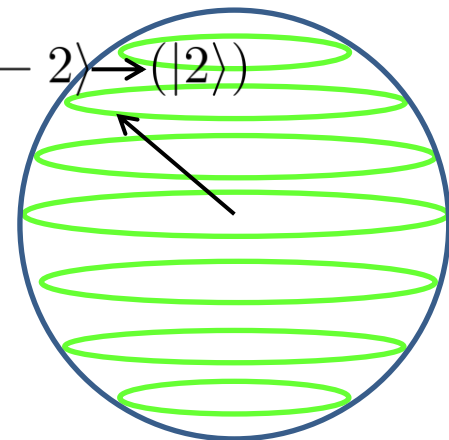
This is as if we are treating number states $|n\rangle$.

Let us introduce boson creation/annihilation operators:

“Vacuum” of the boson: $|S\rangle \quad (S_z = S)$

n -boson state: $|S - n\rangle$

$$\left\{ \begin{array}{l} a_j |n_j\rangle = \sqrt{n_j} |n_j - 1\rangle \\ a_j^\dagger |n_j\rangle = \sqrt{n_j + 1} |n_j + 1\rangle \end{array} \right.$$



Then as is for ordinary boson operators, we obtain:

$$a |S\rangle = 0, \quad |S - n\rangle = \frac{1}{\sqrt{n!}} (a^\dagger)^n |S\rangle$$

With number operator $\hat{n} = a^\dagger a$ we can write

$$\left. \begin{aligned} S_z &= S - \hat{n}, \\ S_+ &= \sqrt{2S - \hat{n}} a, \\ S_- &= a^\dagger \sqrt{2S - \hat{n}} \end{aligned} \right\}$$

Holstein-Primakoff transformation

Non-interacting spin wave approximation

In Holstein-Primakoff transformation we have nonlinear terms. → Interaction between the bosons.

Expand the square roots in Holstein-Primakoff transformation

$$\left. \begin{aligned} \hat{S}_{j+} &= \sqrt{2S} \left(1 - \frac{a_j^\dagger a_j}{4S} + \dots \right) a_j, \\ \hat{S}_{j-} &= \sqrt{2S} a_j^\dagger \left(1 - \frac{a_j^\dagger a_j}{4S} + \dots \right) \end{aligned} \right\}$$

Then the Hamiltonian is expanded as

$$\begin{aligned} \mathcal{H} &= -2 \sum_{\langle i,j \rangle} J_{ij} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j = -2 \sum_{\langle i,j \rangle} J_{ij} \{ \hat{S}_{iz} \hat{S}_{jz} + (\hat{S}_{i+} \hat{S}_{j-} + \hat{S}_{i-} \hat{S}_{j+})/2 \} \\ &= -2 \sum_{\langle i,j \rangle} J_{ij} \left[S^2 - S(\hat{n}_i + \hat{n}_j) + S(a_i^\dagger a_j + a_j^\dagger a_i) + \hat{n}_i \hat{n}_j - \frac{1}{4} a_i^\dagger a_j^\dagger a_j a_j - \frac{1}{4} a_j^\dagger a_j^\dagger a_j a_i + \dots \right]. \end{aligned}$$

Take up to quadratic terms

$$\mathcal{H} = -2 \sum_{\langle i,j \rangle} J_{ij} [S^2 - S(\hat{n}_i + \hat{n}_j) + S(a_i^\dagger a_j + a_j^\dagger a_i)]$$

Ferromagnetic spin wave: ferromagnetic magnon

Fourier transform of creation/annihilation operators:

$$\left. \begin{aligned} a_{\mathbf{q}} &= \frac{1}{\sqrt{N}} \sum_j a_j \exp(i\mathbf{q} \cdot \mathbf{r}), \\ a_{\mathbf{q}}^\dagger &= \frac{1}{\sqrt{N}} \sum_j a_j \exp(-i\mathbf{q} \cdot \mathbf{r}) \end{aligned} \right\}$$

Substitute these to the approximated Hamiltonian \longrightarrow

Magnon Hamiltonian

$$\begin{aligned} \mathcal{H} &= -2 \sum_{\langle i,j \rangle} J_{ij} S^2 + 2 \sum_{\mathbf{q}} [J_0 - J_{\mathbf{q}}] S a_{\mathbf{q}}^\dagger a_{\mathbf{q}} \\ &= E_0 + \sum_{\mathbf{q}} \hbar \omega_{\mathbf{q}} a_{\mathbf{q}}^\dagger a_{\mathbf{q}} \end{aligned}$$

Total magnetization: $M = \mu \left\langle \sum_i S_{iz} \right\rangle = \mu S N - \mu \sum_i \langle a_i^\dagger a_i \rangle = \mu S N - \mu \sum_{\mathbf{q}} n(\epsilon_{\mathbf{q}})$

with Bose distribution function: $n(\epsilon) = \left(\exp \frac{\epsilon}{k_B T} - 1 \right)^{-1}$

Magnon dispersion

$$\hbar \epsilon_{\mathbf{q}} = 2S(J_0 - J_{\mathbf{q}}) = 2SJ \{ 2 - [\exp(iqa) + \exp(-iqa)] \} \simeq 2SJ \left[2 - 2 \left(1 - \frac{(qa)^2}{2} \right) \right] = \underline{2SJ(qa)^2}$$

Then we obtain $M = \mu N \left[S - \zeta \left(\frac{3}{2} \right) \left(\frac{k_B T}{8\pi JS} \right)^{3/2} \right]$ $\zeta(z) = \sum_{n=1}^{\infty} \frac{1}{n^z}$ $\zeta \left(\frac{3}{2} \right) \approx 2.612$

Why we have introduced the concept: Magnon?

Low temperature Magnetic moment: $M = \mu N \left[S - \zeta \left(\frac{3}{2} \right) \left(\frac{k_B T}{8\pi JS} \right)^{3/2} \right]$ $\zeta(z) = \sum_{n=1}^{\infty} \frac{1}{n^2}$ $\zeta \left(\frac{3}{2} \right) \approx 2.612$

The internal energy is: $U = E_0 + \sum_{\mathbf{q}} n(\epsilon_{\mathbf{q}}) = E_0 + 12\pi JSN \zeta \left(\frac{5}{2} \right) \left(\frac{k_B T}{8\pi JS} \right)^{5/2}$.

Then low temperature specific heat is obtained by $C = \frac{\partial U}{\partial T} = \frac{15}{4} N k_B \zeta \left(\frac{5}{2} \right) \left(\frac{k_B T}{8\pi JS} \right)^{3/2}$

As above, by considering magnons we can calculate low energy excitations and obtain important quantities.

Magnons: Low temperature model of ferro (anti-ferro) magnets.

Spin wave modeling of anti-ferromagnets

Anti-ferromagnet → Decompose into A, B sublattices

A sublattice: we can consider magnon model

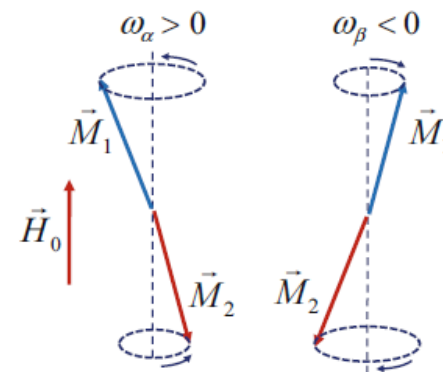
B sublattice: Magnetization is reversed

Take the vacuum as $|0\rangle_B = |-S\rangle$

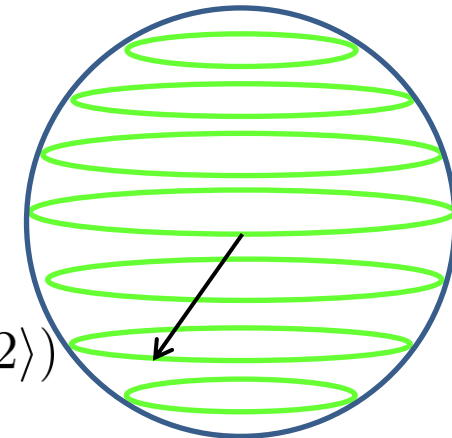
Boson creation/annihilation operators b_j^\dagger, b_j

Then the Holstein-Primakoff transform is

$$\left. \begin{aligned} S_{jz} &= -S + b_j^\dagger b_j, \\ S_{j+} &= b_j^\dagger \sqrt{2S - b_j^\dagger b_j}, \\ S_{j-} &= \sqrt{2S - b_j^\dagger b_j} b_j \end{aligned} \right\}$$



$|-S + 2\rangle \rightarrow (|2\rangle)$



$|-S\rangle \rightarrow (|0\rangle)$

vacuum

Quadratic Hamiltonian : $\mathcal{H} = -\alpha_z |J| N S^2 + 2|J| S \sum_{\langle i,j \rangle} (a_i^\dagger a_i + b_j^\dagger b_j + a_i b_j + a_i^\dagger b_j^\dagger) \quad i \in A, j \in B$

Spin wave modeling of anti-ferromagnets (2)

Fourier transformation of
creation/annihilation operators

$$\left. \begin{aligned} a_i &= \sqrt{\frac{2}{N}} \sum_{\mathbf{q}} a_{\mathbf{q}} \exp(-i\mathbf{q} \cdot \mathbf{r}_i), \\ b_j &= \sqrt{\frac{2}{N}} \sum_{\mathbf{q}} b_{\mathbf{q}} \exp(-i\mathbf{q} \cdot \mathbf{r}_j) \end{aligned} \right\}$$

Momentum representation of
the Hamiltonian

$$\mathcal{H} = -\alpha_z |J| N S^2 + 2\alpha_z |J| S \sum_{\mathbf{q}} [a_{\mathbf{q}}^\dagger a_{\mathbf{q}} + b_{\mathbf{q}}^\dagger b_{\mathbf{q}} + \gamma(\mathbf{q})(a_{\mathbf{q}}^\dagger b_{\mathbf{q}}^\dagger + a_{\mathbf{q}} b_{\mathbf{q}})]$$

with nearest neighbor summation

$$\gamma(\mathbf{q}) = \alpha_z^{-1} \sum_{\rho} \exp(-i\mathbf{q} \cdot \rho)$$

But the above Hamiltonian is still not diagonalized. (Néel ordered state is not true ground state.)

Bogoluibov transformation

$$(a_{\mathbf{q}}, b_{\mathbf{q}}) \rightarrow (\alpha_{\mathbf{q}}, \beta_{\mathbf{q}})$$

$$\left. \begin{aligned} a_{\mathbf{q}} &= \cosh \theta_{\mathbf{q}} \alpha_{\mathbf{q}} - \sinh \theta_{\mathbf{q}} \beta_{\mathbf{q}}^\dagger, \\ b_{\mathbf{q}} &= \cosh \theta_{\mathbf{q}} \beta_{\mathbf{q}} - \sinh \theta_{\mathbf{q}} \alpha_{\mathbf{q}}^\dagger. \end{aligned} \right\}$$

Bosonic commutation relations

$$[\alpha_{\mathbf{q}}, \alpha_{\mathbf{q}}^\dagger] = 1, \quad [\beta_{\mathbf{q}}, \beta_{\mathbf{q}}^\dagger] = 1, \quad [\alpha_{\mathbf{q}}, \beta_{\mathbf{q}}] = [\alpha_{\mathbf{q}}^\dagger, \beta_{\mathbf{q}}^\dagger] = 0$$

Spin wave modeling of anti-ferromagnets (3)

$$\mathcal{H} = -\alpha_z |J| N S^2 + 2\alpha_z |J| S \sum_{\mathbf{q}} [(\cosh 2\theta_{\mathbf{q}} - \gamma(\mathbf{q}) \sinh \theta_{\mathbf{q}})(\alpha_{\mathbf{q}}^\dagger \alpha_{\mathbf{q}} + \beta_{\mathbf{q}}^\dagger \beta_{\mathbf{q}} + 1) - 1 - (\sinh 2\theta_{\mathbf{q}} - \gamma(\mathbf{q}) \cosh 2\theta_{\mathbf{q}})(\alpha_{\mathbf{q}} \beta_{\mathbf{q}} + \alpha_{\mathbf{q}}^\dagger \beta_{\mathbf{q}}^\dagger)]$$

Condition for diagonalization: $\sinh 2\theta_{\mathbf{q}} / \cosh 2\theta_{\mathbf{q}} = \tanh 2\theta_{\mathbf{q}} = \gamma(\mathbf{q})$

Diagonalized Hamiltonian: $\mathcal{H} = -\alpha_z |J| N S^2 + 2\alpha_z |J| S \sum_{\mathbf{q}} [(\sqrt{1 - \gamma(\mathbf{q})^2} - 1) + \sqrt{1 - \gamma(\mathbf{q})^2}(\alpha_{\mathbf{q}}^\dagger \alpha_{\mathbf{q}} + \beta_{\mathbf{q}}^\dagger \beta_{\mathbf{q}})]$

Ground state energy: $-\alpha_z |J| N S^2 + 2\alpha_z |J| S \sum_{\mathbf{q}} [(\sqrt{1 - \gamma(\mathbf{q})^2} - 1)]$

Néel ordered state energy Energy lowering due to hybridization of $S, S - 1, \dots, -S$ states



$\langle S_{jz} \rangle$ is shorter than S

How? $\langle S_{jz} \rangle = S - \frac{2}{N} \sum_{\mathbf{q}} \sinh \theta_{\mathbf{q}} = S - \frac{1}{N} \sum_{\mathbf{q}} \left(\frac{1}{\sqrt{1 - \gamma(\mathbf{q})^2}} - 1 \right)$

Spin wave modeling of anti-ferromagnets (4)

$$\langle S_{jz} \rangle = S - \Delta \quad E_0 = N|J|\alpha_z S(S + \epsilon)$$

Lattice	Square	Simple Cubic	Body Centered Cubic
Δ	0.917	0.078	0.0593
ϵ	$0.158 + 0.0062S^{-1}$	$0.097 + 0.0024S^{-1}$	$0.073 + 0.0013S^{-1}$

Magnon dispersion $\epsilon_{\mathbf{q}} = 2\alpha_z |J| S \sqrt{1 - \gamma(\mathbf{q})^2}$ Simple cubic case $\gamma(\mathbf{q}) = \cos \frac{q_x}{2} \cos \frac{q_y}{2} \cos \frac{q_z}{2}$

Asymptotic form $q \rightarrow 0$ $\epsilon_{\mathbf{q}} = 2\sqrt{2\alpha_z} |J| S a q$

Internal energy $U = E_0 + \frac{\pi^2}{15} N \left(\frac{k_B T}{2\sqrt{2\alpha_z} |J| S} \right)^3 k_B T$

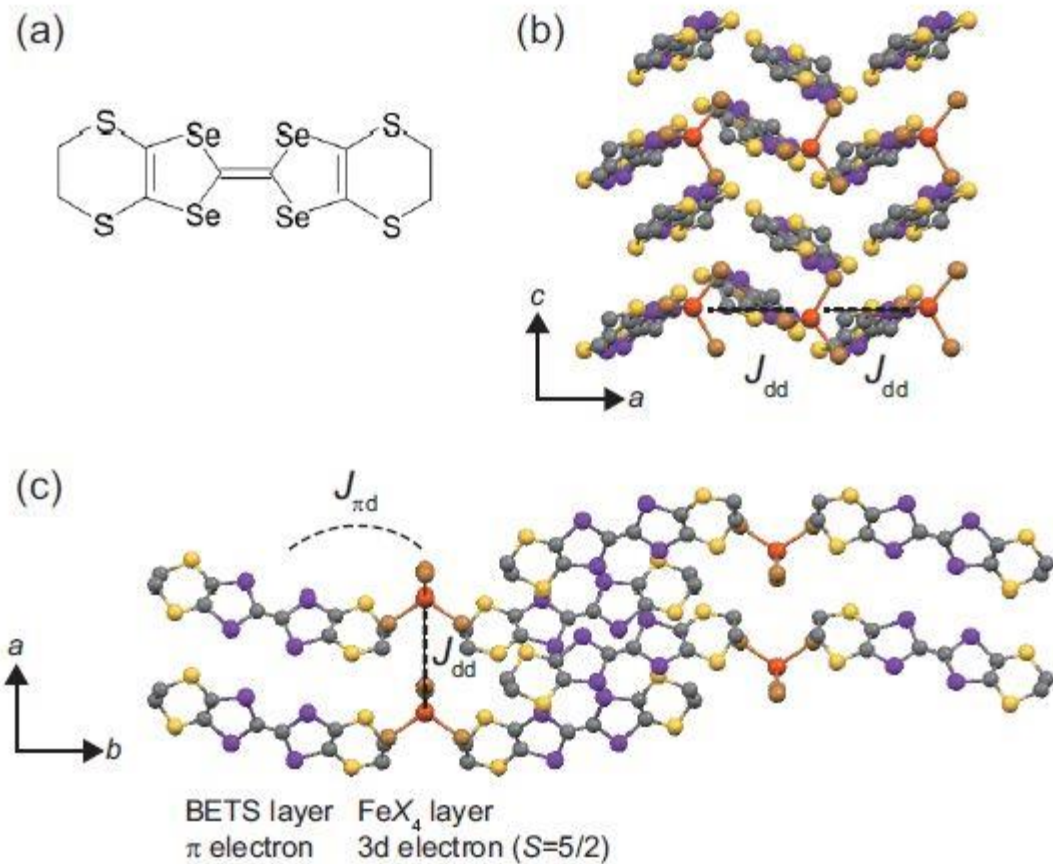
Lattice	1D Chain	2D Square Lattice	3D Simple Cubic
$-\frac{E_0}{\alpha_z J N S^2}$	$1 + 0.363S^{-1}$	$1 + 0.158S^{-1}$	$1 + 0.097S^{-1}$
$\frac{C}{Nk_B}$	$\frac{2\pi}{3} \left(\frac{k_B T}{2\alpha_z J S} \right)$	$\frac{14.42}{\pi} \left(\frac{k_B T}{2\alpha_z J S} \right)^2$	$4\sqrt{3} \frac{\pi^2}{5} \left(\frac{k_B T}{2\alpha_z J S} \right)^3$
ΔS	Diverge	0.197	0.078

Specific heat of an organic anti-ferromagnet

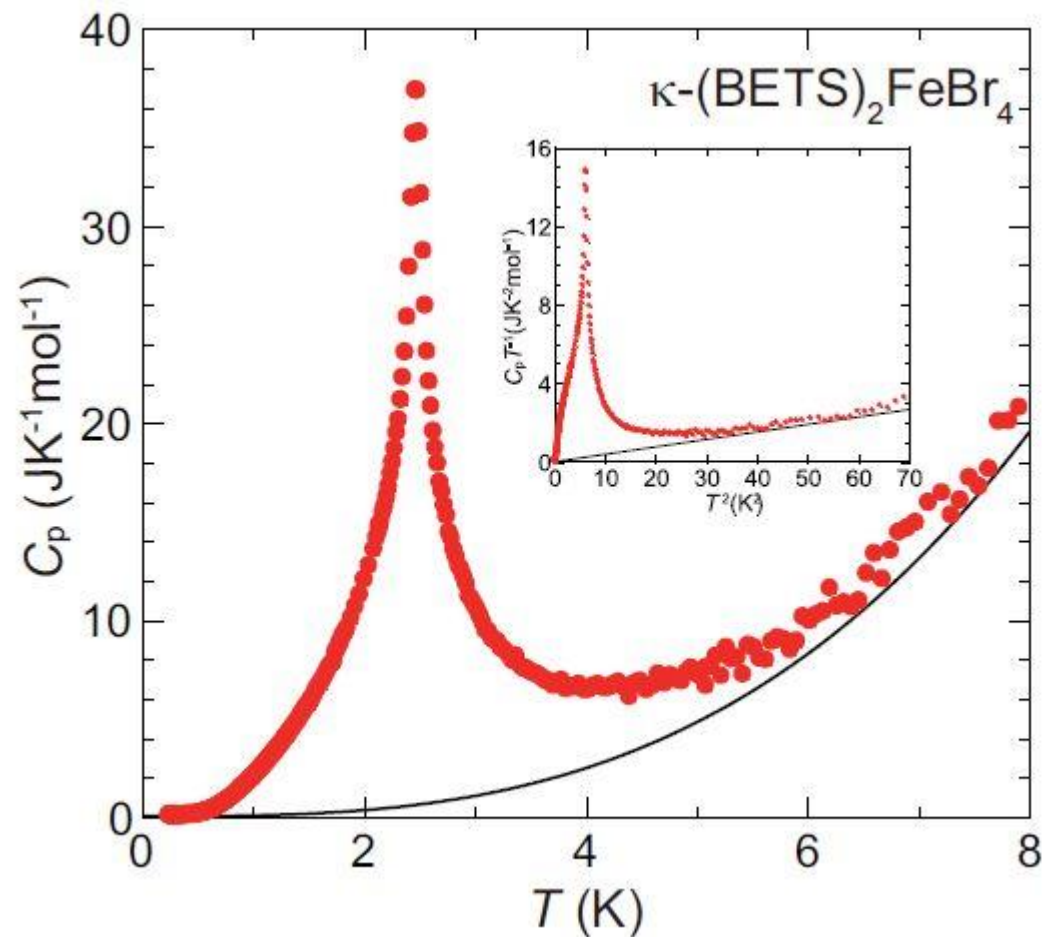


Anti-ferromagnetic T_N : 2.5 K

Superconducting T_c : 1.1 K

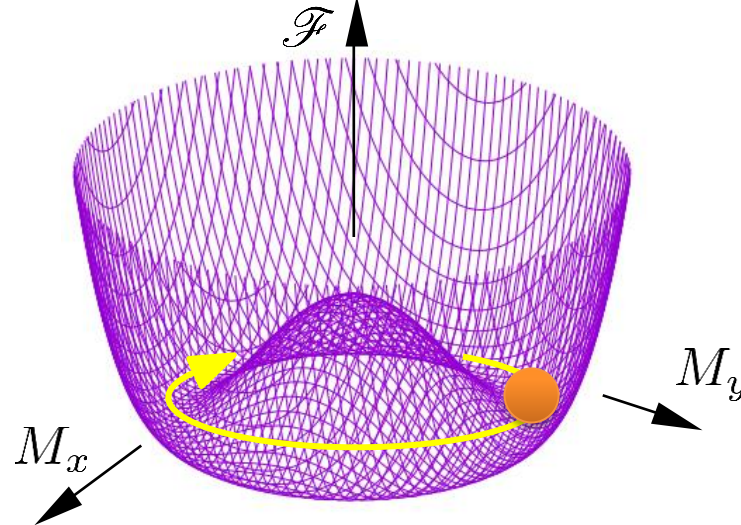
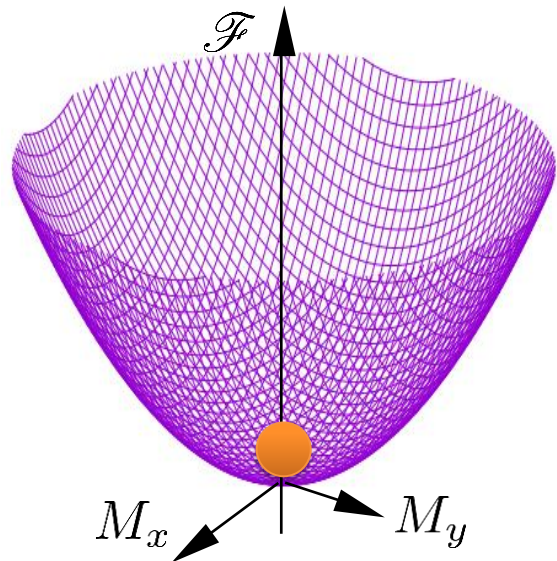
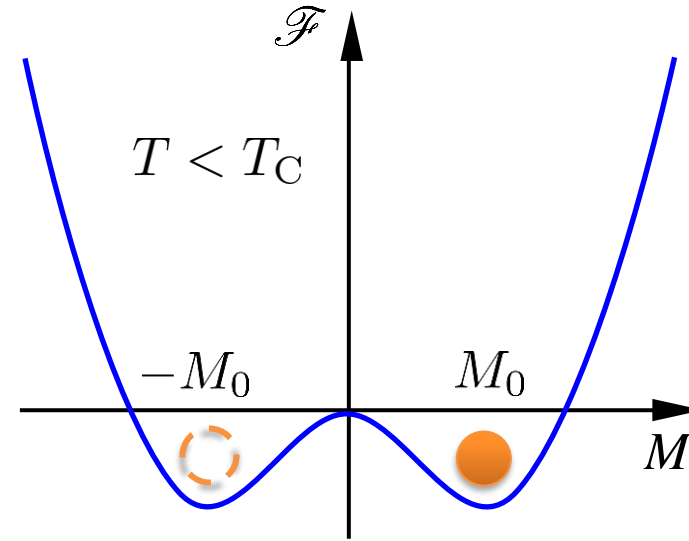
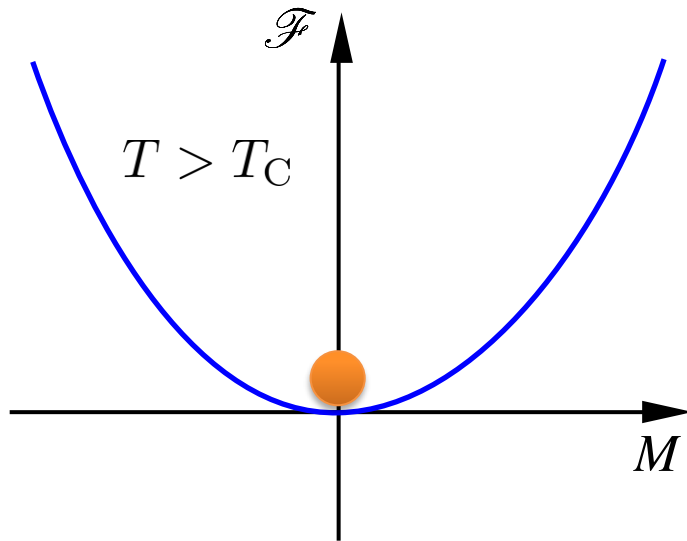


Normal metal specific heat: $C_m = AT + BT^3$



Fukuoka et al., PRB **93**, 245136 (2016)

Spontaneous Symmetry Breaking and Nambu-Goldstone mode



Spontaneous symmetry breaking

Nambu-Goldstone theorem

When a spontaneous symmetry breaking takes place, a mode with zero energy at long wavelength limit appears.

$$E = mc^2$$

$$\begin{matrix} \downarrow & \downarrow \\ 0 & 0 \end{matrix} \text{ massless}$$

Nambu-Goldstone mode
(Nambu-Goldstone boson)

Magnons in the case of ferromagnets (type-B) and anti-ferromagnets (type-A).

Generalization of Nambu-Goldstone theorem (column)

Nambu-Goldstone mode, Higgs mode → Birth of particle mass; Standard theory of elementary particles
The theories based on the principle prevail all over the physics.

However, there still have been many open questions!

An example: According to the primitive statement, the number of NG mode should be the same as that of broken symmetries.

	Broken symmetry	Number of NG modes	Number of broken symmetry
N-G theorem		x	$y = x$
Crystal	Translational symmetry	3	3
3D Ferromagnet	Rotational symmetry	1	2
Spinor BEC	Rotational symmetry	2	3
Skymion crystal	Translational symmetry	1	2

Extended theorem (2012) : $x = y - \text{rank} \langle [Q_a, Q_b] \rangle / 2$

Magnon dispersion relation measurement in MnF₂

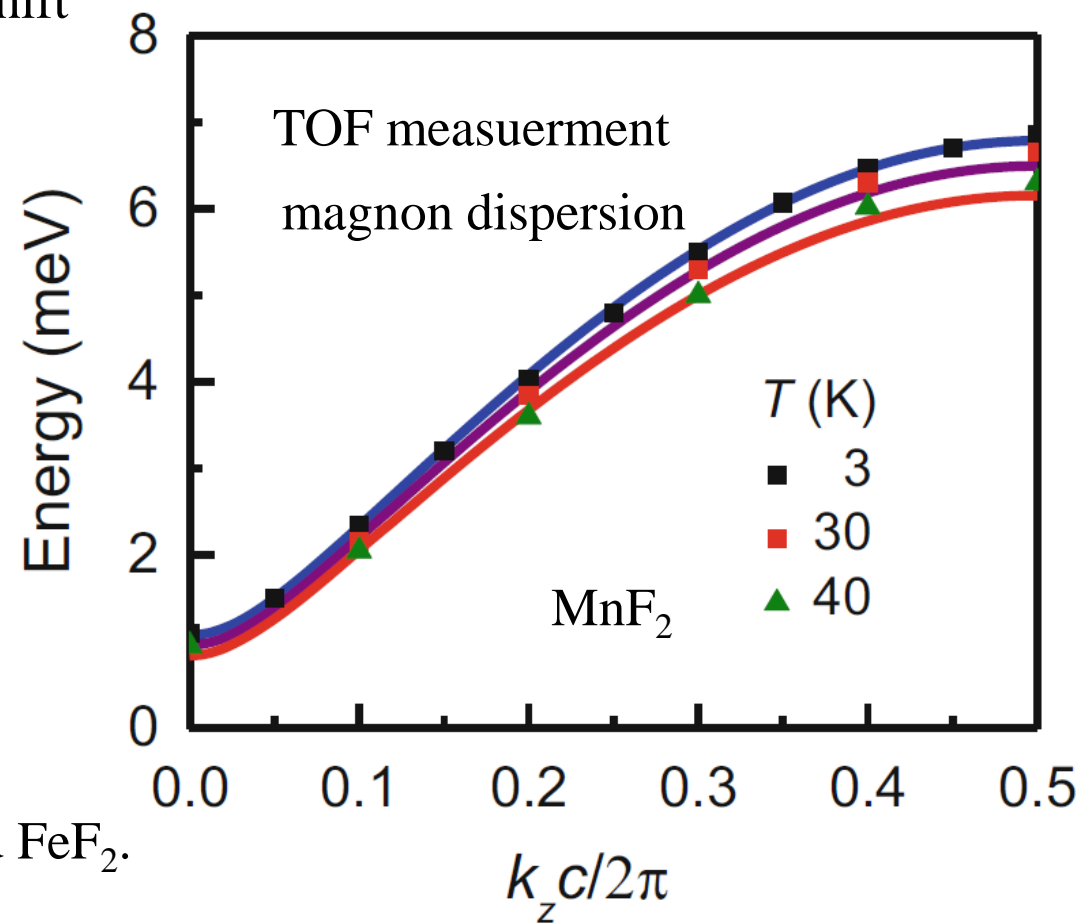
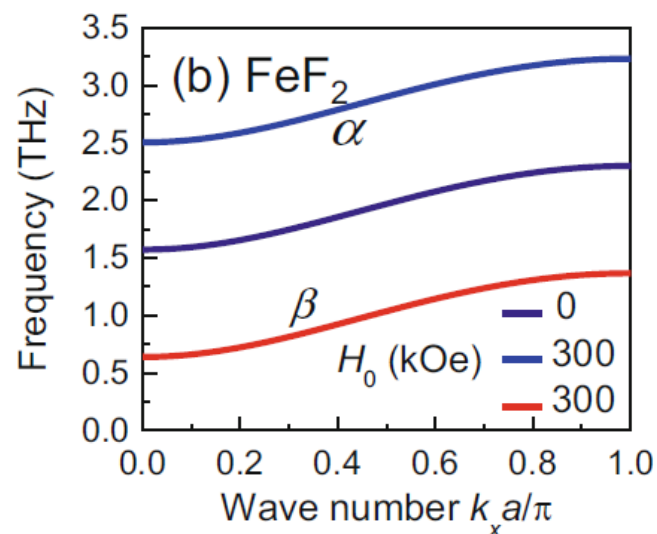
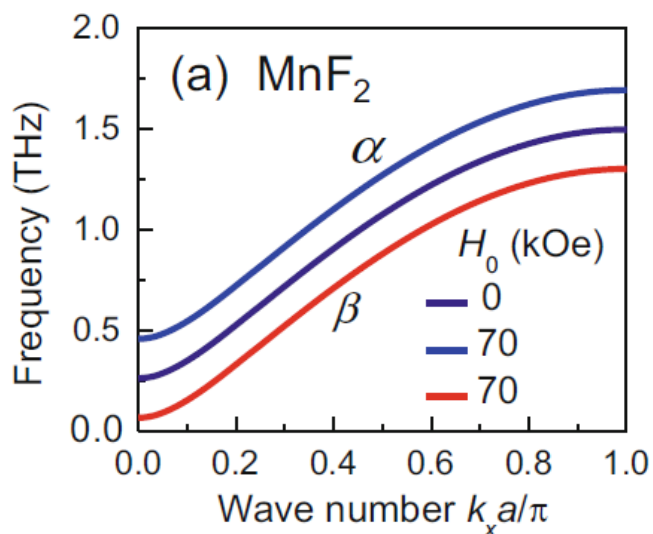
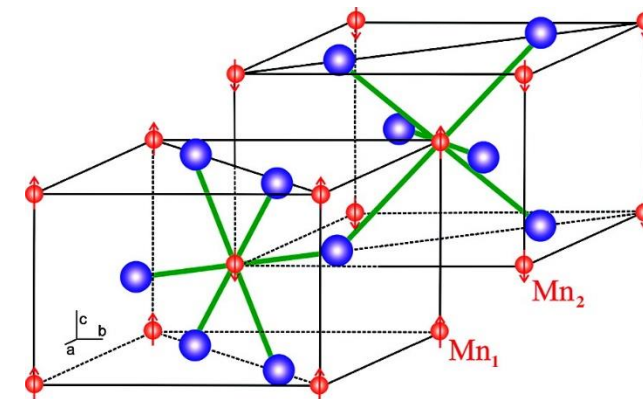
Neutron time-of-flight (TOF) measurement

$$\mathcal{H}_{\text{int}} = -\boldsymbol{\mu}_e \cdot \mathbf{B}_n \quad \mathbf{B}_n = \text{rot} \left(\boldsymbol{\mu}_n \times \frac{\mathbf{r}}{r^3} \right) \quad \text{neutron magnetic field}$$

electron moment

neutron inelastic scattering

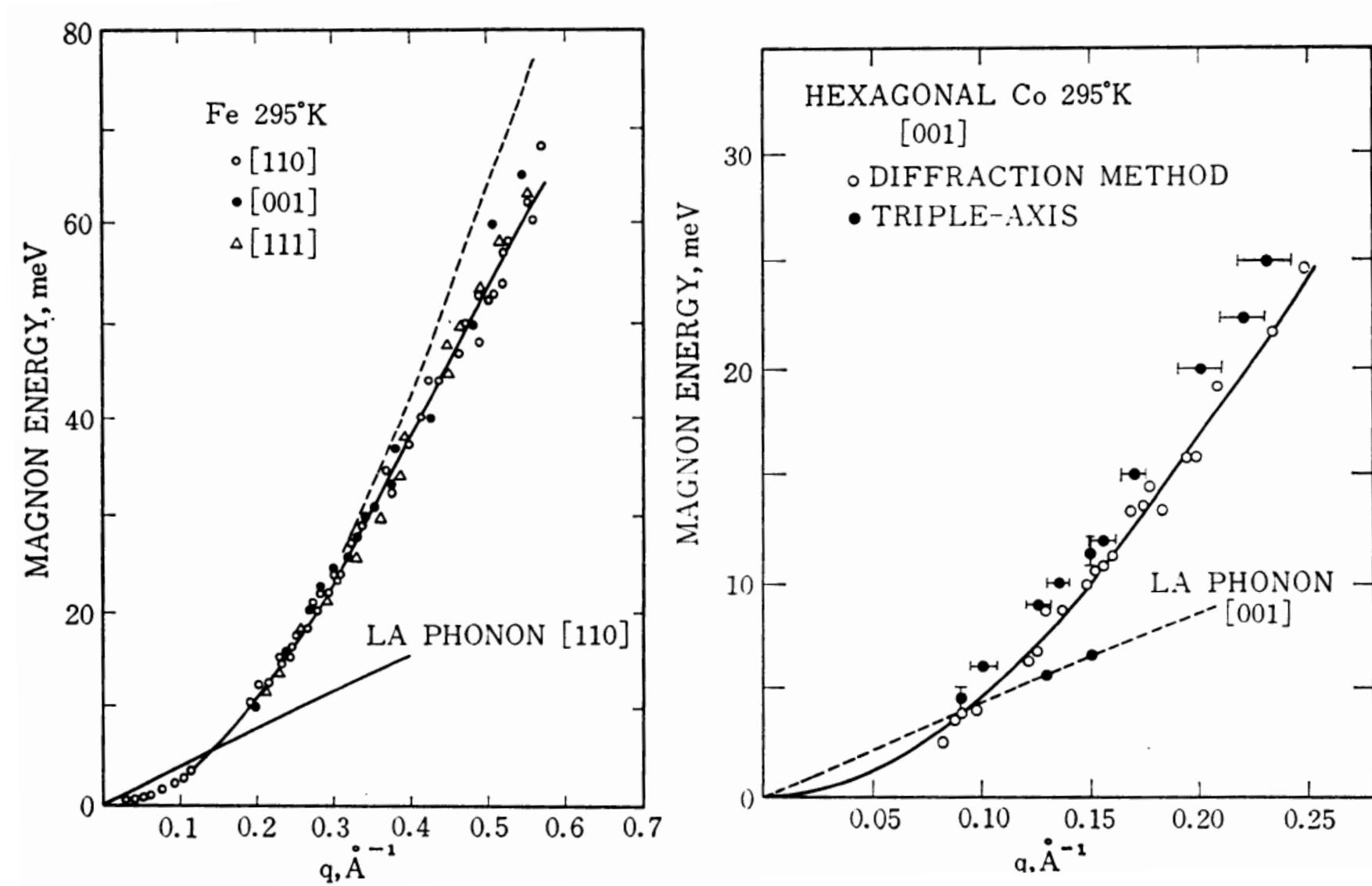
$$\left\{ \begin{array}{l} \hbar\mathbf{k} \longrightarrow \hbar(\mathbf{k} - \mathbf{q}) \quad \text{momentum shift} \\ \Delta E = \frac{\hbar^2}{2M} (-2\mathbf{k} \cdot \mathbf{q} + \mathbf{q}^2) \quad \text{energy loss} \end{array} \right.$$



Calculated dispersions of anti-ferromagnetic magnons in MnF₂ and FeF₂.

(Taken from *Fundamentals of Magnonics*.)

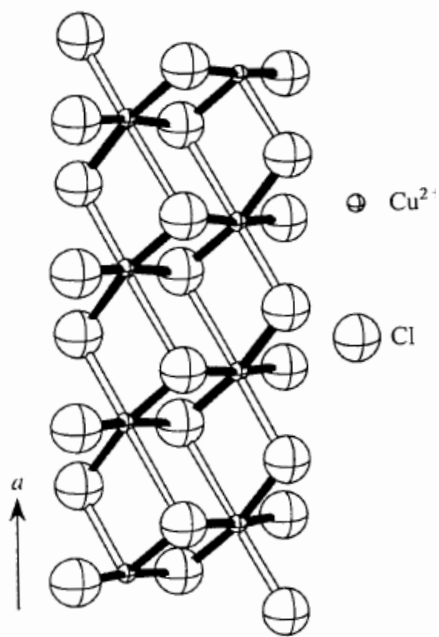
Magnon dispersions in metallic ferromagnets



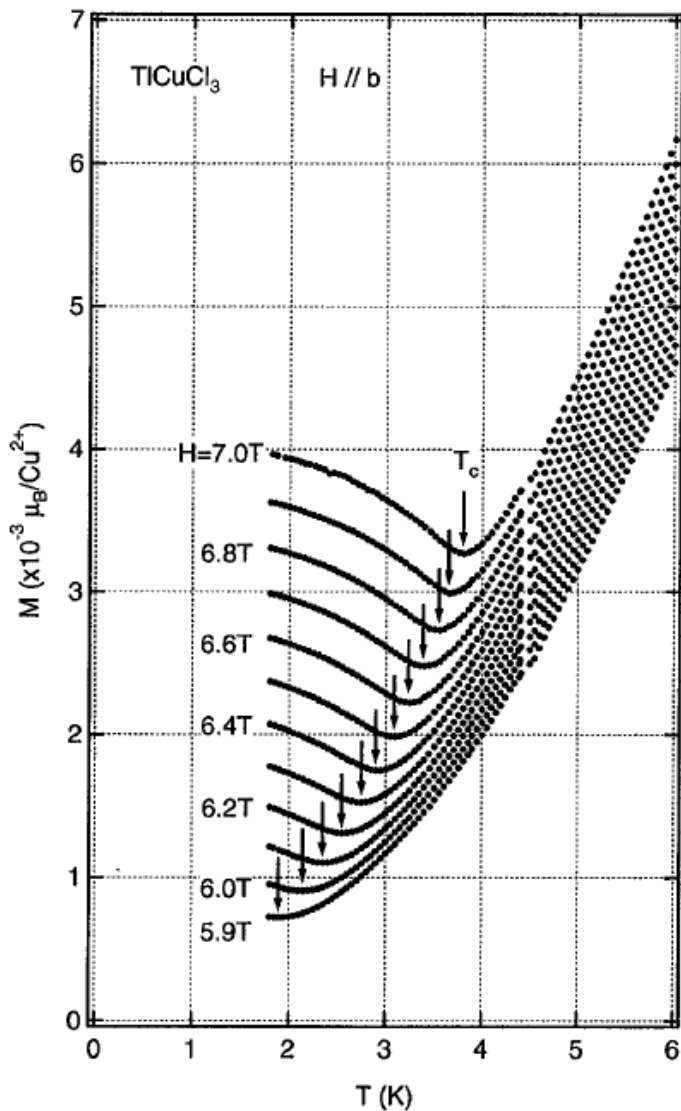
Bose-Einstein condensation of magnons

Magnons: not completely bosons (para statistics) however can be treated as “hard core” bosons

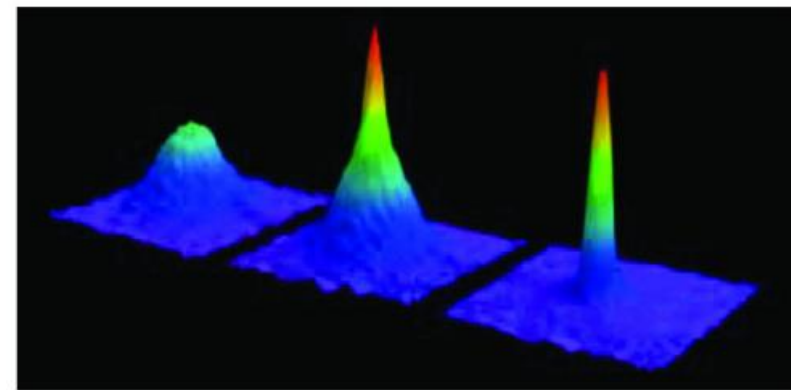
TlCuCl_3



Nikuni et al.,
PRL **84**, 5868
(2000)

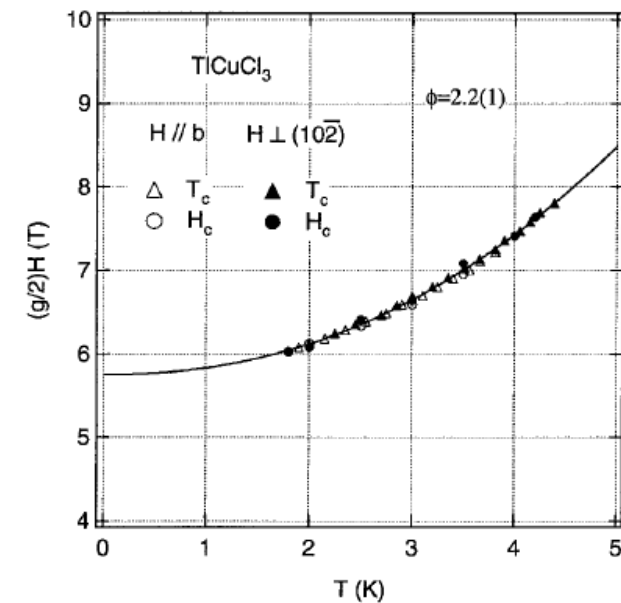
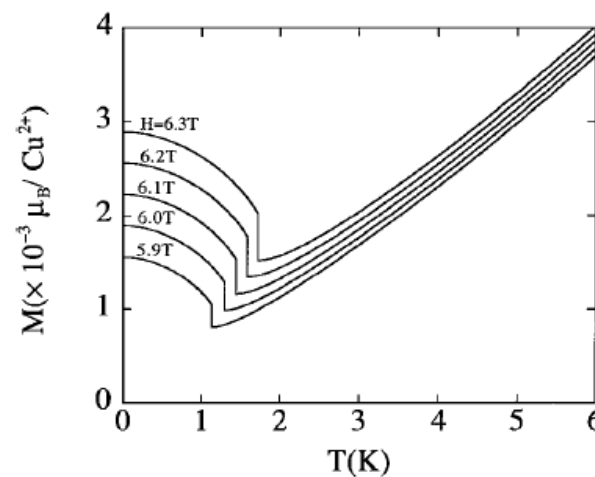


Sharp enhancement of magnetization

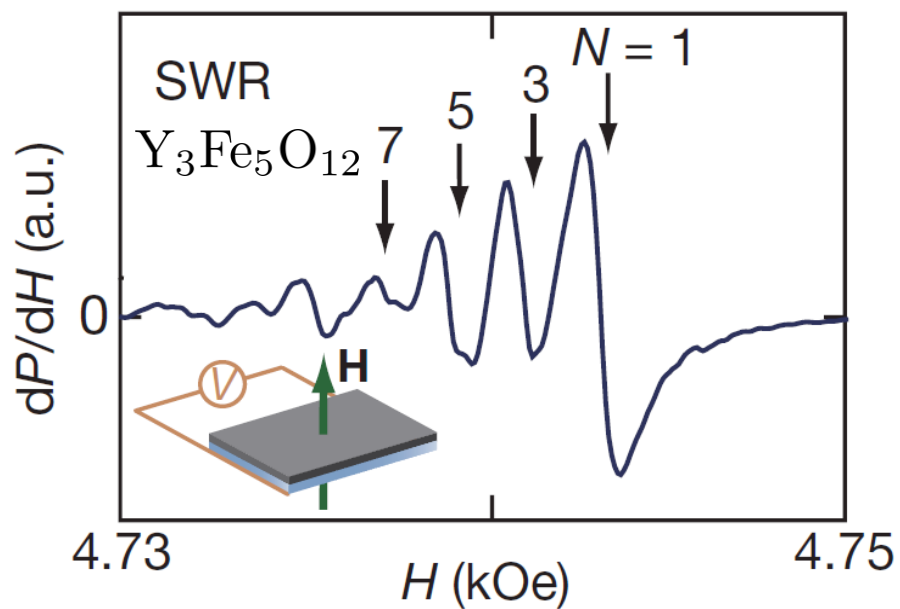


BEC in cold Rb atom ensemble
Sharp increase in particle density

Theory



Magnon (spin wave) resonance in thin films



Kajiwara et al., Nature **464**, 262 (2010)

Spin wave resonance in a thin film

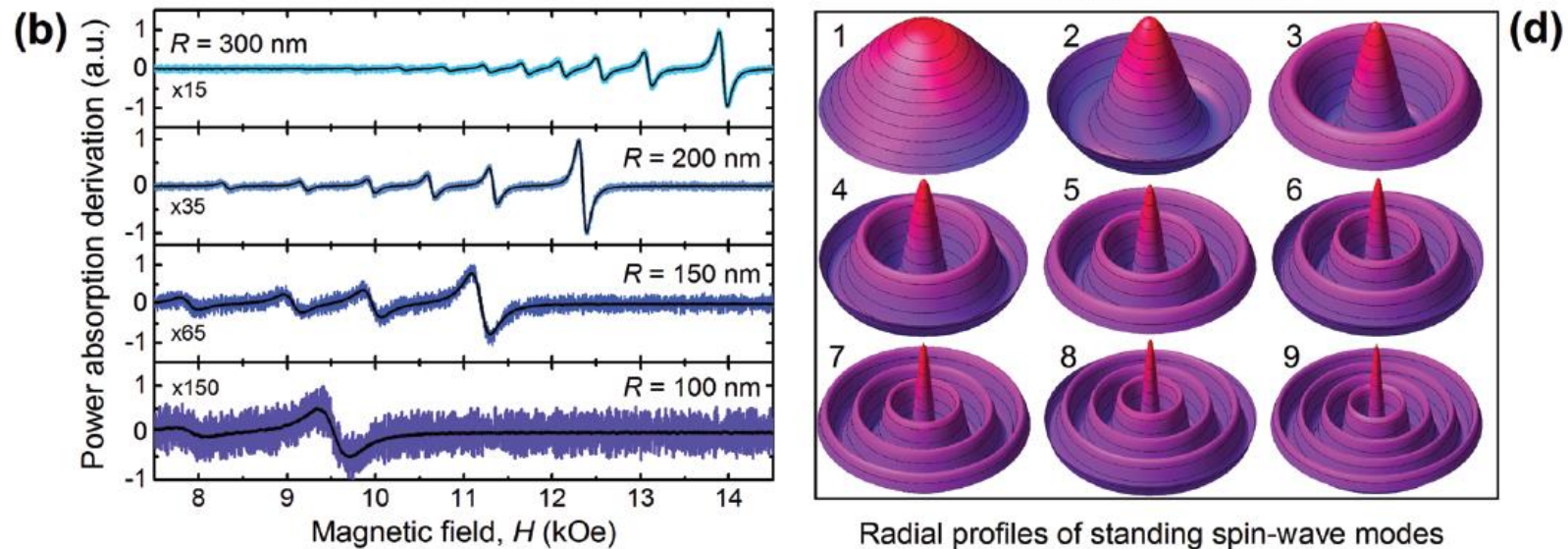
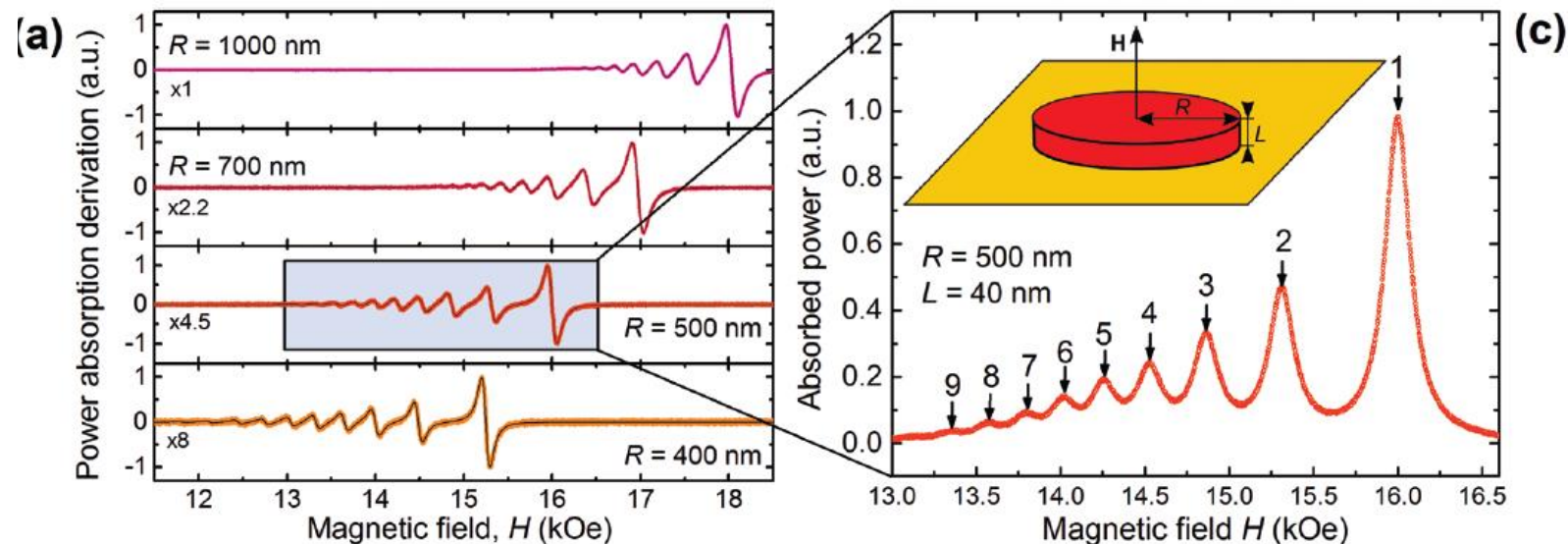
$$\frac{m}{\gamma} = \frac{B}{\mu_0} - 4\pi M_S + \frac{2Ak^2}{M_S}$$

$$k = \frac{p\pi}{L} \quad p : \text{odd number,}$$

$$L : \text{film thickness} \quad A = \frac{\alpha_z S^2 J}{a}$$

Kittel Phys. Rev. **110**, 1295 (1958)

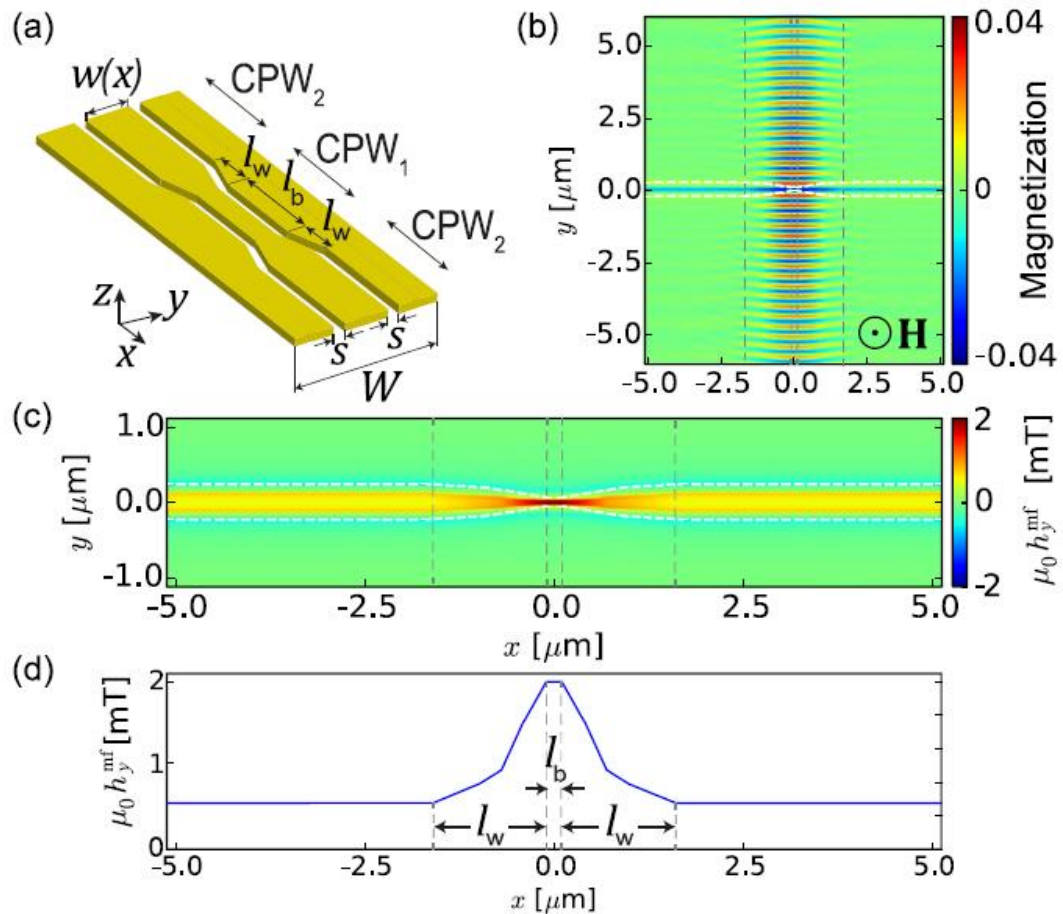
CoFe Nano disk



Dobrovolskiy et al., Nanoscale **12**, 21207 (2020).

Real space imaging of magnons

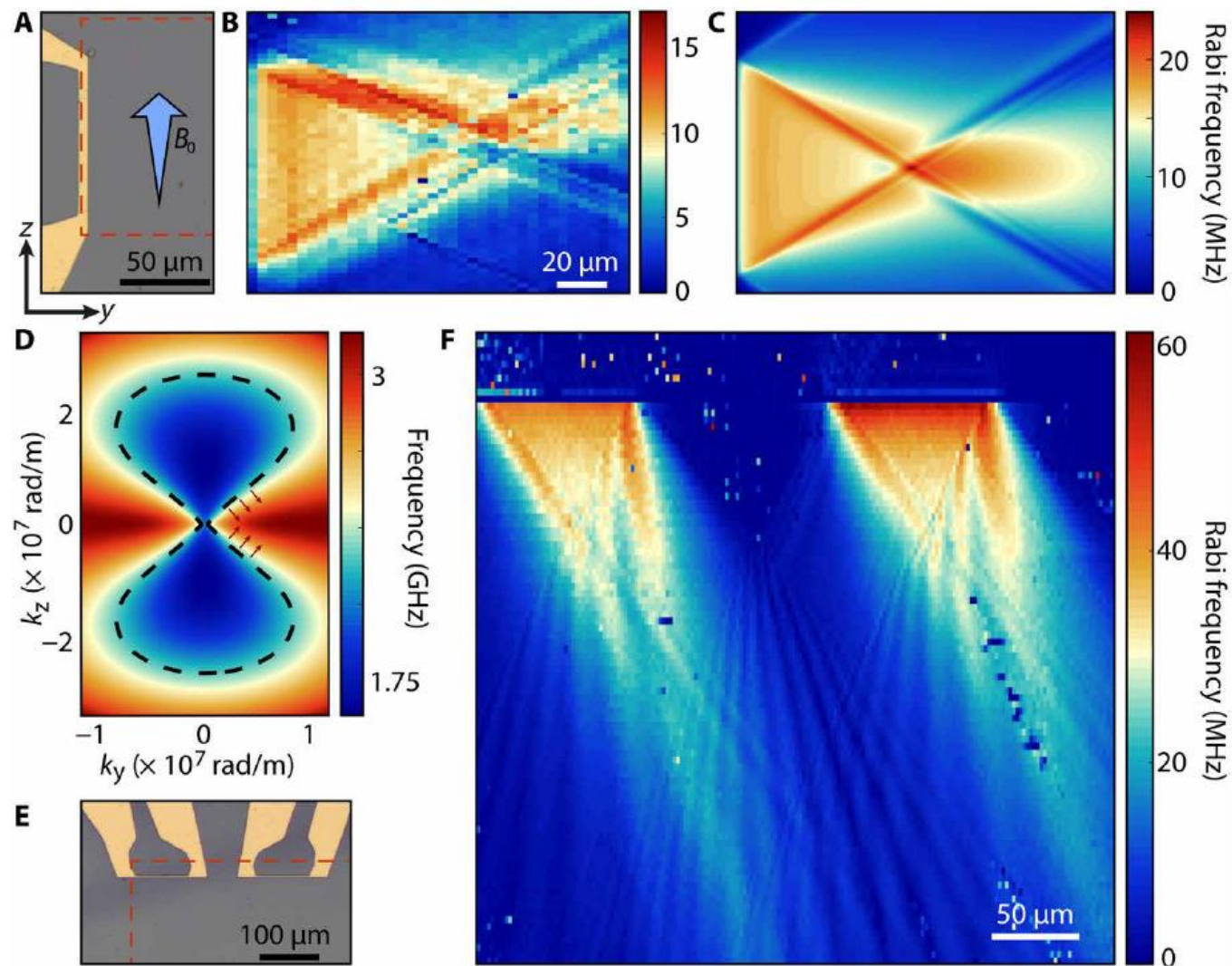
YIG



Gruszecki et al. Sci. Rep. **6**, 22367 (2016)

YIG

NV center ESR detection of magnon fields



Bertelli et al., Sci. Adv. **6**, eabd3556 (2020).

Summary

- Spin wave (classical) in a ferromagnet
- Quantization of spin wave (magnon)
- Magnons in an anti-ferromagnet
- Magnon approximation for weak excitations in ferro- and anti-ferromagnets
- Nambu-Goldstone mode in phase transition
- Experiments on magnons