Nuclear Magnetic Relaxation Time near Compensation Temperature in Ferrimagnetic Insulator

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The nuclear magnetic relaxation time $T_1$ in ferrimagnetic insulators is calculated by a Raman process of hyperfine interaction with a meanfield approximation. It is found that the $1/T_1$ on one site rapidly increases near the compensation temperature $T_c$, whereas that on another site does not increase up to Curie temperature $T_c$. This is due to that the band width of soft magnon becomes comparable to $T_c$. The increasing behavior of $1/T_1$ below $T_c$ is found also in another type ferrimagnet, which shows hump structure in the temperature dependence of magnetization instead of compensation. Also in this case, we find the rapid increase of $1/T_1$ below $T_c$, even though the magnetization does not show the compensation. Such a coexistence of soft and hard magnons will lead to remarkable properties of ferrimagnet.

1. Introduction

The ferri-magnet is a kind of ferro-magnet and was theoretically predicted by Néel. Soon after, the magnetization compensation was observed in the LiFeCr spinel ferrite, whose magnetization becomes zero at magnetization compensation temperature $T_M$ far below Curie temperature $T_c$. Such a ferrimagnet is called N-type and was also found in rare-earth iron garnets (RIGs). Those were studied by many authors in order to apply their magnetization compensation to a magneto-optical memory.

Dynamical aspects of ferrimagnetism were initially studied by the electron spin resonance (ESR). Different from the ferromagnetic resonance (FMR), the ferrimagnetic resonance (FIR) has two branches. One gives the usual FMR, while another one is called exchange frequency and is located higher in energy. It was difficult to measure the exchange frequency at that time, since its wavelength was of the order of a tenth of a millimeter. However, a singular behavior of gyro-magnetic ratio was observed around $T_M$ in LiFeCr spinel ferrite. Regarding the lower branch, an effective gyro-magnetic ratio becomes small around $T_M$ and then rapidly increases around the angular momentum compensation temperature $T_A$. The $g$-value of upper branch becomes small into a measurable range around $T_A$. A magnetization is a product of Lande’s $g$-factor and a total angular momentum. In general, hence, $T_M$ is different from $T_A$, when an orbital angular momentum is active. In contrast to the magnetization, the dynamics of ferrimagnet becomes singular around $T_A$.

It should be noted that a magnetization couples to a magnetic field, while an total angular momentum itself does not. So that it might be difficult to directly measure $T_A$ using conventional methods. Recently, however, Imai et al. has successfully observed $T_A$ using Barnet effect. On a rotating frame, its rotational frequency is couples to an angular momentum instead of magnetization without any coupling constant. By the spin-rotation coupling, a magnetization is induced through an angular momentum by a mechanical rotation. This was originally studied by Barnet and is now used to specify $T_A$ in RIG. It is reported that, around $T_A$, a magnetization reversal becomes quick and a domain wall moves fast. Those properties advantageous for magnetic memory are attributed to the angular momentum compensation.

The nuclear magnetic resonance (NMR) is also powerful tool for magnetism of broad materials. Magnetic excitations can be characterized by the nuclear magnetic relaxation time $T_1$ whose origin is the hyperfine interaction between electron and nucleus. Regarding magnetic insulators, however, the origin of $T_1$ is not so obvious. If the system is isotropic and the quantization axes of nuclear and electron are identical, the relaxation cannot be obtained within the linearized spin-wave approximation. The misalignment of quantization axes and/or the dipole-dipole interactions between an electronic and a nuclear spins will induce the relaxation called Raman process. Those interactions among magnons are also the source of relaxation, e.g., three magnon process. Those are studied for a ferrimagnetic and an antiferromagnetic insulators. Recently, Imai et al. have reported an enhancement of NMR signal around $T_A$, which is closely related to the domain wall motion. In contrast to the ESR, the NMR is a site-selective measurement of magnetism. It must be interesting to study dynamical aspect of magnetism in ferrimagnet site by site. In addition, consistent understanding of ferrimagnetism among experimental methods, NMR, ESR, and neutron scattering will be useful.

In this paper, we will study the nuclear magnetic relaxation time in the ferrimagnet. In Sec. 2, a model Hamiltonian and an approximation will be explained. The nuclear magnetic relaxation time by the Raman process will be given in Sec. 3. Additional changes due to an orbital angular momentum will be discussed in Sec. 4. Below, Bohr magneton $\mu_B$ and Planck constant $\hbar = h/2\pi$ are set to 1 for brevity.

2. Formalism: Magnons in Ferrimagnet

We will focus on a ferrimagnetic "insulator", which is simply called "ferrimagnet" below. The magnetic exchange interaction by the Pauli principle and Coulomb interaction between electrons is the source of magnetism in a ferrimagnet. Two sub-lattice with different magnitudes of spins $S_A \neq S_B$ is the simplest model. The Hamiltonian is given by,

$$H = -J_A \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j - J_B \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j + J_C \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j.$$ (1)
with spin operators $\vec{S}_i$ on site $i$ in A-site ($i$ in B-site). The magnitudes of magnetic exchange interactions $J_A$, $J_B$, and $J_C$ are supposed to be positive for brevity. First, we do not consider an orbital angular momentum $\vec{L}_i$. Hence, there is only one compensation temperature $T_0$, i.e., $T_M = T_C = T_0$. What is changed by $\vec{L}_i$ will be discussed in the last section. At $T_0$, the expectation values $(S_i^A \equiv M_A > 0$ and $(S_i^B \equiv -M_B < 0$ satisfy $M_A - M_B = 0$, where the bracket denotes the thermal average. See also Appendix A. It is known that there are some possible cases of compensation. In the above one, both sub-lattices have same number of sites in a unit cell as shown in the inset of Fig. 1. Another one is $M_{A}A = M_{B}B = 0$, when the number of sub-lattice $A$, $n_A$, is different from that of sub-lattice $B$, $n_B$. As shown in Appendix B, the characteristic features will be common among those lattice structures. Hence, we consider the simplest one shown in Fig. 1 below.

The compensation occurs at finite temperatures. To include the temperature dependences of $M_A$ and $M_B$, we adopt the meanfield approximation and the linearized spinwave approximation around the meanfield solution. This is equivalent to the Tyablikov decoupling in the Greens function method and is a kind of random phase approximation.\(^{37,38}\) The meanfield solution for $J_A=0.1$, $J_B=1.0$, $J_C=0.05$, $S_A=1$, and $S_B=1/2$, is shown in Fig. 1, and $T_0/T_c \sim 0.3$ with the Curie temperature $T_c$ ($\sim 3.0$).

The Holstein–Primakov (HP) bosons (magnons) $a_i^\dagger$, $a_i$ on A-sublattice and $b_j^\dagger$, $b_j$ on B-sublattice are given by, $S_i^- \sim \sqrt{2} M_A a_i^\dagger$, $S_i^+ \sim \sqrt{2} M_A a_i$, $S_j^- = M_A - a_i^\dagger a_i$, $S_j^+ \sim \sqrt{2} M_B b_j^\dagger$, $S_j^- \sim \sqrt{2} M_B b_j$, $S_j^+ = b_j^\dagger b_j - M_B$. Below, the spins are supposed to be ordered in $z$-direction. By the linearized approximation, the action of magnons is given by.\(^{39}\)

$$ S = \sum_{q,\omega_n} \Phi^\dagger \left( \begin{array}{cc} -i \omega_n & 0 \\ 0 & i \omega_n \end{array} \right) \left( \begin{array}{c} \epsilon_{1q} \\ \epsilon_{2q} \end{array} \right) \Phi, \quad (2)$$

$$ \Phi^\dagger \equiv (a_i(i \omega_n), b_j(-i \omega_n)), \quad (3)$$

$\epsilon_{1q} \equiv z J_C M_B + z_A J_A M_A (1 - \zeta_{1q})$, $\epsilon_{2q} \equiv z J_C M_A + z_B J_B M_B (1 - \zeta_{2q})$, $\epsilon_{3q} \equiv z J_C \sqrt{M_A M_B} \gamma_q$, $\zeta_{A(B)} \equiv \frac{1}{z_{A(B)}} \sum_n \cos(q_n)$, $\gamma_q \equiv \frac{1}{\epsilon} \sum_{n \neq 0} e^{i \eta_n}$, by using the boson operators $a_i(i \omega_n)$ and $b_j(-i \omega_n)$ with momentum $q$ and Matsubara frequency for bosons $\omega_n$. The dispersion relation of magnon depends on a connectivity of sub-lattice giving the number of nearest neighbor sites among each sub-lattice, $z_A$ and $z_B$, and that of nearest neighbor sites between two sub-lattices, $z$. In Eqs. (7) and (8), $\eta$ means the summation about the nearest neighbor sites among each sub-lattice and between two sub-lattices. By Eq. (2), the Green functions of magnons $g_{A}(q, i \omega_n)$ and $g_{B}(q, i \omega_n)$ are obtained as,

$$ g_{A}(q, i \omega_n) \equiv \frac{1}{\xi_{1q}} + \frac{\xi_{2q}}{(i \omega_n - E_{1q})}, \quad (9)$$

$$ g_{B}(q, i \omega_n) \equiv \frac{1}{\xi_{3q}} + \frac{\epsilon_{2q}}{(i \omega_n - E_{2q})}, \quad (10)$$

with the dispersion relation of magnons $E_{aq}$ and $E_{bq}$ given by,$$ E_{aq} = \frac{1}{2} \left( \xi_{1q} - \xi_{2q} \right) + \frac{1}{2} \left( \epsilon_{1q} - \epsilon_{2q} \right) - \frac{1}{2} \left[ (\xi_{1q} + \epsilon_{2q})^2 - 4 \xi_{3q} \right], \quad (11)$$

$$ E_{bq} = \frac{1}{2} \left( \xi_{1q} - \xi_{2q} \right) + \frac{1}{2} \left( \epsilon_{1q} - \epsilon_{2q} \right) - \frac{1}{2} \left[ (\epsilon_{1q} + \epsilon_{2q})^2 - 4 \xi_{3q} \right]. \quad (12)$$

The small $q$ approximation for $M_A > M_B$ leads to,$$ E_{aq} \sim C q^2, \quad (13)$$

$$ E_{bq} \sim 12J_C (M_A - M_B) + D q^2, \quad (14)$$

where $C$ and $D$ are constants given in the Appendix B and $q = \sum_{i=x,y,z} q_i^2$. One mode $E_{aq}$ gapless, while the other one $E_{bq}$ has the "optical gap", $E_{aq} \equiv |E_{aq}| = 12J_C (M_A - M_B)$, which disappears at $T_0$. Then, the dispersion relations degenerate at the gamma point and linearly increases with $q$ similar to the antiferromagnet. Away from the gamma point, on the other hand, the two dispersion relation deviate from each another as,$$ E_{aq} \sim C_1 |q| + C_2 q^2, \quad (15)$$

$$ E_{bq} \sim C_1 |q| - C_2 q^2. \quad (16)$$

where $C_1$ and $C_2$ are constants given in the Appendix B. Those $q$-dependences will be relevant to the temperature dependence of $T_1$ at low temperatures.

### 3. Results: Nuclear Magnetic Relaxation by Raman Process

In this study, we consider the nuclear magnetic relaxation time $T_1$ originating from the contact interaction between a nu-
clear and an electron given by,
\[
H_{n-el} = \frac{1}{2} \sum_{\nu=\alpha,\beta} \left[ g_{n} \sum_{i} f_{\nu i} \left( S_i^z \cdot I_i^z \right) \right],
\]
with $g$-factor on $\nu$-site $g_{\nu}$. If the system is isotropic and the quantization axes of nuclear and electron are identical, we cannot obtain the relaxation within the linearized approximation. The misalignment of quantization axes and/or the dipole-dipole interactions between an electronic and a nuclear spins will induce the relaxation called Raman process.\(^{32-34}\) The interactions among magnons are also the source of relaxation, e.g., three magnon process.\(^{32-34}\) Below, we will focus on the Raman process induced by the misalignment. This will be enough to find some characteristics of $T_1$ near $T_0$. A critical exponent with respect to $T$ of $T_1$ is beyond the scope of this study and will be discussed elsewhere. When the quantization axis of nucleus is deviated by $\theta$ in angle from that of electron, the following components are reduced from Eq. (17),
\[
H^c_{n-el} = \frac{1}{2} \sum_{\nu=\alpha,\beta} \left[ g_{\nu} \sum_{i} \sin \theta f_{\nu i} \left( I_i^z + I_i^z \right) \right],
\]
which are relevant to calculate $T_1$ and $g_{\nu} = g_{\alpha} = g$. Assumed that the form factors $f_{\alpha \nu}$ are constant as, $f_{\alpha \nu} = f_{\beta \nu} \equiv f$, the $T_1$ on site $\nu = A, B$ is given by,
\[
\frac{1}{T_{1\nu}} = F \sum_{q} C_{\nu}(q, \omega_0),
\]
where $\langle \cdots \rangle$ means the thermal average. The nuclear magnetic resonance energy is denoted by $\omega_0$ and $F = (g_{\nu} f \sin \theta/2)^2$. Using Eqs. (9) and (10), the spin-spin correlation function $C_{\nu}(q, \omega_0)$ is described by,
\[
C_{\nu}(q, \omega_0) = \frac{2}{1 - \cos(q/k_B T)} \text{Im} \Pi^\nu_{\nu}(q, \omega_0),
\]
\[
\Pi_{\nu}(q, i\omega_0) = k_B T \sum_{k,\omega} g_{\nu}(k + q, \omega_0; \nu \omega_0) g_{\nu}(k, \omega_0),
\]
with temperature $T$ and the Boltzmann constant $k_B$. The retarded function of $\Pi_{\nu}(q, i\omega_0)$ is denoted by $\Pi^R_{\nu}(q, \omega_0)$. When $\omega_0$ is much smaller than $k_B T$, the nuclear magnetic relaxation on site $\nu$, $T_{1\nu}$, is given by,
\[
\frac{1}{T_{1\nu}} = 2 F \sum_{q \neq 0} \left| \frac{\text{Im} \Pi^R_{\nu}(q, \omega_0)}{q} \right|,
\]
\[
\frac{1}{T_{1\nu}} = 2 \pi F \sum_{q \neq 0} \left| n_{B}(E_{\nuq}) \right| \left| n_{B}(E_{\nuq}) + 1 \right| \left[ \frac{\alpha_{\nu} \alpha_{\nu}}{\Delta_{\nu}} \delta(E_{\nuq} - E_{\nuq}) + n_{B}(E_{\nuq}) \right],
\]
\[
\alpha_{\nu} = \frac{1}{2} \left( \frac{E_{p1} + E_{p2}}{\Delta_{\nu}} + 1 \right),
\]
\[
\beta_{\nu} = \frac{1}{2} \left( \frac{E_{p1} + E_{p2}}{\Delta_{\nu}} - 1 \right),
\]
\[
\Delta_{\nu} = \sqrt{(E_{p1} + E_{p2})^2 - 4|E_{p1}|^2},
\]
in which $\mu \neq \nu$, i.e., $\mu = \alpha$ for $\nu = \beta$ or $\mu = \beta$ for $\nu = \alpha$ and the Bose distribution function is denoted by $n_{B}(\epsilon) \equiv 1/[e^{\epsilon/(k_B T)} - 1]$. Note that Eq. (24) can be checked by considering the ferromagnet and the antiferromagnet as discussed in Appendix C.

Using Eq. (24) and the meanfield solution shown in Fig. 1, the $T$-dependence of $1/T_{1\nu}$ is numerically calculated as shown in Fig. 2 (a). The $1/T_{1A}$ and $1/T_{1B}$ are plotted by upward triangle (red) and downward triangle (blue), respectively. The meanfield solution for $J_A=0.1$, $J_B=1.0$, $J_C=0.05$, $S_A=1$, and $S_B=1/2$ are used. The broken line indicates $T_0$. See also Fig. 1. (b) At low temperatures, $T_{1A}$ is well fitted by $T^2$ (broken line) similar to the ferromagnet (See also Appendix C), whereas it is deviated by increasing temperature as $AT^2 + BT^3$ (solid line). $A$ and $B$ are some constants.

![Fig. 2](image-url)

(a) The $T$-dependence of $1/T_1$ on site-A and site-B (1/T_{1A} and 1/T_{1B}) are plotted by upward triangle (red) and downward triangle (blue), respectively. The meanfield solution for $J_A=0.1$, $J_B=1.0$, $J_C=0.05$, $S_A=1$, and $S_B=1/2$ in Fig. 1. It is noted that 1/T_{1A} rapidly increases around $T/T_0 \sim 0.3$, that corresponds to $T_0$ indicated by the broken line in Fig. 2. This is sharp contrast to 1/T_{1B}, which diverges just below $T_c$. As shown in Fig. 2 (b), at low temperatures, $T_{1A}$ is well fitted by $T^2$ similar to the ferromagnet (See also Appendix C). By increasing temperature, on the other hand, it is fitted by $AT^2 + BT^3$ with some constants $A$ and $B$. This is similar behavior of ferromagnet except for the fact that 1/T_{1A} increases around $T_0$ instead of $T_c$.

To understand this behavior of 1/T_{1A} around $T_0$, $E_{pA}$ and $E_{pB}$ are plotted in Fig. 3 for (a) $T/T_c = 0.1$ and (b) $T/T_c = 0.3$ with...
$q_x = q_z = 0$. At low temperatures, the $T$-dependence of $1/T_1$ is determined by $q^2$-dependences of $E_{qA}$ and $E_{qB}$ around $q \sim 0$. See also the inset of Fig. 3 (a). At $T = T_0$, $E_q$ becomes zero as shown in the inset of Fig. 3 (b), and both $E_{qA}$ and $E_{qB}$ become proportional to $q$ instead of $q^2$ around $q = 0$. Note that $k_BT/J_B$ is shown by the broken line as measure of temperature in Figs. 3 (a) and (b). In the preset lattice structure, $E_{qA}$ is maximum at $\vec{q} = (\pi, \pi/2, 0)$. It can be approximated at $\vec{q} = (\pi, 0, 0)$, since we choose a small value of $J_C = 0.05$. Then, we find that, at $T = T_0$, the band width of $E_{qA}$ becomes comparable to $k_BT$. It means that all states of $E_{qA}$ contribute to $1/T_1$ through $n_{qA}(x)$ of the first term in Eq. (24), where the first term is dominant. This is the origin of rapidly increasing behavior of $1/T_1A$ at $T_0$. It was not accidental due to the followings. The band width $E_{qA}$ is very roughly estimated by, $\varepsilon_{qA} \approx (6J_C+16J_A)A_0 \sim 8J_A = 0.8$ with $M_A = M_B \equiv M_0 \sim 0.5$. On the other hand, $T_0$ is roughly estimated by, $T_0 = \gamma_A J_A X_A = 0.8$. Since $M_A$ is soft and rapidly decreases with $T$, $T_0$ is close to the Curie temperature of the system limited to $A$-sublattice. Therefore, $1/T_1A$ rapidly increases around $T_0$.

So far, we have discussed a ferrimagnet called $N$-type. Another type of ferrimagnet called $P$-type shows a hump in a temperature dependence of magnetization instead of the compensation. Figure 4 is calculated by Eq. (1) for $J_A=0.5$, $J_B=1.0$, $J_C=0.2$, $S_A=1/2$, and $S_B=1$. The lattice structure is same to the inset of Fig. 1. In the $P$-type, the magnetization does not show any singular behavior such as compensation, although it is composed of two different sublattices, i.e., soft and hard such as $E_{qA}$ and $E_{qB}$. The $1/T_1$ in the $P$-type ferrimagnet is plotted in Fig. 5 in the same way of $N$-type. We find that $1/T_1A$ rapidly increases around $T/T_C \sim 0.4$, close to the top of the hump structure, while $1/T_{1B}$ does near $T_C$. It is now straightforward to understand this behavior, since $A$-sublattice is soft and $B$-sublattice is hard. It is clear to see the dispersion relation shown in Figs. 6 for (a) $T/T_C = 0.1$ and (b) $T/T_C = 0.4$. In each figures, $k_BT/J_B$ is shown by the broken line as measure of temperature. At $T/T_C = 0.4$, all of $E_{qA}$ contribute to $1/T_1A$. Therefore, even in the $P$-type ferrimagnet without compensation, we will find a rapid increase of $1/T_1$.

4. Summary and Discussions

We have studied $T_1$ in the ferrimagnetic insulator induced by the Raman process of hyperfine interaction. To calculate $1/T_1$, the Heisenberg model composed of two sublattices is adopted and the linearized spinwave approximation around the meanfield solution is used. At $T_0 < T_c$, $1/T_1$ on one sub-
landé $g$-factor on R-site is different from that on iron site. So that $T_M \neq T_A$ in general. Still, Eq. (1) is our starting point. The expectation value of $S_z^\alpha$ has an extra factor (ga-1) such as $\langle S_z^\alpha \rangle = (ga-1) \langle f_x^\alpha \rangle$.\(^{(40)}\) Those factors can be renormalized into $J_A$, $J_B$, and $J_C$ as $K_A \equiv (ga-1)^2 J_A$, $K_B \equiv (ga-1)^2 J_B$, and $K_C \equiv (ga-1)(gb-1) J_C$.$^{(41,42)}$ Using $K_A$, $K_B$, and $K_C$, the dispersion relation of magnons are obtained by substituting $\langle f_x^\alpha \rangle$ and $\langle f_x^\beta \rangle$ for $M_A$ and $M_B$, respectively. See also Appendix D. Around which temperature, $T_M$ or $T_A$, does $1/T_1$ start to increase? The band width of magnon is determined by the expectation value of $\langle S_z^\alpha \rangle = (ga-1) \langle f_x^\alpha \rangle$ and $\langle S_z^\beta \rangle = (gb-1) \langle f_x^\beta \rangle$ instead of $\langle f_x^\alpha \rangle$ and $\langle f_x^\beta \rangle$.  It is reminded that $1/T_1$ increases, when $k_B T$ is comparable to the band width, and $T_A$ is determined by $\langle f_x^\alpha \rangle$ and $\langle f_x^\beta \rangle$. For example, in the case of $ga=5/4$, $gb=2$ such as Ho$_3$Fe$_5$O$_{12}$, the factors $(ga-1)$ and $(gb-1)$ are smaller than 1. Roughly estimated, hence, the energy scale of band width will be smaller than $T_A$. It means that $1/T_1$ will start to increase further blow $T_A$. However, those energy scales are different depending on materials. So that it is difficult to identify the temperature, at which $1/T_1$ starts to increase. Such a material dependence will be discussed in the near future and will be clarified experimentally.

On the other hand, it is clear that $E_g$ becomes zero at $T_A$ instead of $T_M$.

$$E_g = 12 K_C \left( \langle f_x^\alpha \rangle - \langle f_x^\beta \rangle \right). \quad (28)$$

The magnon excitations in RIGs are reported by inelastic neutron scattering.$^{35,36}$ However, $E_g$ around the compensation temperatures has not yet been clarified. The loss of $E_g$ must be associated with the increase of domain wall speed at $T_A^{28-31}$ and the enhancement of NMR signal.$^{25}$ Such remarkable changes of domain wall will make the ferrimagnet more useful material for spintronics. More important is that consistent understanding among NMR, ESR, and neutron scattering will be useful.

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### Appendix A: Meanfield Equation

The meanfield equation and its solution are straight forward.

$$M_A = fS_A \left[ (S_A J_A M_A - \varepsilon JC M_B) / (k_B T) \right] , \quad (A-1)$$

$$M_B = fS_B \left[ (S_B J_B M_B - \varepsilon JC M_A) / (k_B T) \right] , \quad (A-2)$$

$$fS [x] \equiv (S + 1/2) \coth [x(S + 1/2)] - 1/2 \coth (x/2) . \quad (A-3)$$

Note that $g$ and $\mu_B$ are abbreviated for clarity of the equations. Expanding Eqs. (A-1) and (A-2), $T_c$ is given by,

$$T_c = \frac{1}{2} \left[ X_A S_A J_A + X_B S_B J_B \right] + \sqrt{ \left( X_A S_A J_A - X_B S_B J_B \right)^2 + X_A X_B (2zJ_C)^2} . \quad (A-4)$$

$$X_A \equiv (S_A + 1) S_A / 3 , \quad (A-5)$$
Appendix B: Dispersion Relation of Magnons in Ferromagnet

At low energies away from the compensation temperature, the dispersion relation of magnons Eqs. (11) and (12) are expanded as,

\[ E_{\alpha\beta} \sim Cq^2, \]

\[ E_{\beta\eta} \sim 12JC(M_\alpha - M_\beta) + Dq^2, \]

\[ C = \frac{[3J_3M_\alpha^2 - (J_A + J_B - 2JC)M_AM_B + 3J_6M_B^2]}{M_A - M_B}, \]

\[ D = \frac{J_4M_\alpha^2 - (3J_3 + 3J_6)M_AM_B + J_B^2}{M_A - M_B}. \]

where \( q^2 \equiv q_x^2 + q_y^2 + q_z^2 \). Note that \((0,0)\) and \((\pi, \pi)\) are equivalent in the case of Fig. 1 (a). At the compensation temperature \( M_A = M_B \equiv M \), the excitation gap vanishes,

\[ E_{\alpha\beta} \sim Cq^2 + C\beta |q|, \]

\[ E_{\beta\eta} \sim Cq^2 + C\beta |q|, \]

\[ C_1 = 2\sqrt{3}JC(J_A + J_B + J_C)S, \]

\[ C_2 = 2(J_A - J_B)S, \]

\[ \epsilon_{\alpha\eta} + \epsilon_{\beta \eta} = 12JC \sqrt{3}(1 + 4J_6)S q^2, \]

\[ \Delta_\alpha = 6JE(M_A \alpha - M_B \beta), \]

\[ \Delta_\beta = 12JC(M_B \alpha + M_A \beta) + 24J_6 \sqrt{3}M_AM_B q^2, \]

This does not depend on the lattice structure as shown in Fig. B.

Appendix C: Nuclear Magnetic Relaxation in Ferromagnet and Antiferromagnet

The ferromagnetic state is obtained by imposing \( S_B = 0 \) and \( J_C = J_B = 0 \) to the meanfield equation of Eq. (1). Equation (24) is reduced to,

\[ \frac{1}{T_{1,v}} = 2\pi A \sum_{p,q} n_B(E_{vp}) [n_B(E_{vp}) + 1] \frac{[\epsilon_{p\beta} \epsilon_{q\eta}]}{\Delta_p \Delta_q} + \frac{1}{4}, \]

with \( E_{\alpha\beta} = z_\alpha \chi_\alpha M_\alpha (1 - \zeta_{\alpha\beta}) \). The temperature dependence of \( 1/T_{1,v} \) is shown in Fig. C.1. At low temperatures, it is well fitted by \( CT^2 \) due to \( E_q \propto q^2 \). On the other hand, it is deviated from \( T^2 \) by increasing \( T \) and is well fitted by \( AT^2 + BT^5 \), since \( q \)-linear component grows in \( E_q \). Here, \( A, B, C \) are some constants.

The antiferromagnetic state is also done by \( S_A = S_B \) and \( J_A \)

Appendix D: ESR Frequencies

When we consider \( J, J_A, J_B, \) and \( J_C \) are substituted by \( K_A, K_B, \) and \( K_C, \) and further \( M_A \) and \( -M_B \) are interpreted as \( \{ J_A \} \) and \( \{-J_B \} \) in Eqs. (11) and (12). In a magnetic field \( H = (0,0,H) \), \( g_A H \) and \( -g_B H \) are added to Eqs. (4) and (5), respectively. The magnon excitations at \( q = 0 \), which correspond to ESR frequencies \( \Omega_A \) and \( \Omega_B \), are given by,

\[ \Omega_A = \frac{1}{2} \sqrt{\epsilon_1^2 - \epsilon_2^2 + 4|\epsilon_3|^2}, \]
with effective gyromagnetic ratio \( \gamma_{\text{eff}} \). At \( T_A \), it is noted that two frequencies becomes close each another as, \( \Omega_\alpha - \Omega_\beta = (g_A + g_B)H \).

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