Effect of dimensionality crossover on magnetovolume properties of quasi one-dimensional weakly antiferromagnetic metals

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Abstract. Magnetic properties and thermal expansion of quasi one-dimensional itinerant electron magnets close to the antiferromagnetic instability are studied. By controlling the relative spectral dispersions of spin fluctuation spectra depending on the direction of crystal axes, we show that the three dimensional properties are changed with increasing the one-dimensional anisotropy. Crossover behaviors are discussed on the temperature dependence of the magnetic susceptibility and the magnetovolume expansion around their critical regions.

1. Introduction
Since the spin fluctuation theory of magnetovolume effect was published in 1980 by Moriya and Usami [1], the volume change of crystals from the magnetic origin have been usually analyzed by the following formula:

\[ \omega \equiv \frac{\delta V}{V} = \rho KC[M^2(T) + \xi^2(T)], \quad \xi^2(T) = \sum_q < \delta M_q \cdot \delta M_{-q} >, \]  

where \( M(T) \), \( \delta M_q \), and \( \xi(T) \) represent the magnetization, the wave-vector dependent magnetic fluctuation and the thermal spin fluctuation amplitude. The magnetovolume coupling constant, the compressibility, and a density of magnetic atoms are, respectively, denoted by \( C \), \( K \), and \( \rho = N_0/V \), where \( N_0 \) and \( V \) are the number of magnetic atoms and the volume of the crystal. The result was derived by extending the Stoner-Wohlfarth-Edwards free energy [2] of the magnetovolume effect, simply by including the effect of thermal spin fluctuations. Though (1) looks reasonable, it violates the thermodynamic consistency because Grüneisen’s relation between the magnetic specific heat and the thermal expansion coefficient is not satisfied at low temperatures, as was pointed out by Takahashi and Nakano [3].

Recently, the inconsistency was resolved by Takahashi and Nakano [3, 4]. Their thermal volume expansion is derived from the explicit volume dependence of the same free energy that is used in their treatment of the magnetic specific heat. Grüneisen’s relation is, therefore, automatically satisfied at low temperatures. According to them, the thermal expansion in itinerant electron ferromagnets is dominated by the \( T^2 \)-linear behavior at low temperatures, and
the $T$-linear slope of the thermal expansion coefficient rapidly increases towards the magnetic instability point. Similar enhancement of the thermal expansion coefficients has been also predicted [5] for itinerant nearly and weakly antiferromagnetic metals near the instability point.

A recent study on thermal expansion coefficients of lower-dimensional itinerant weak antiferromagnets shows even stronger enhancement around the magnetic instability point [5]. It is, however, very difficult to find strictly low-dimensional materials in nature. Only usually found are quasi two-dimensional or one-dimensional systems. The purpose of this paper is, therefore, to study the magnetic properties and the thermal expansion of quasi one-dimensional exchange enhanced paramagnets (P) and antiferromagnets (AF) close to the instability points. Our particular concerns are the crossover from three-dimensional (3-D) to one-dimensional (1-D) behaviors on the temperature dependence of the above magnetic and magnetovolume properties.

In what follows in section 2, we show how magnetic properties of quasi one-dimensional itinerant magnets close to the antiferromagnetic instability are predominantly determined by the effect of spin fluctuations. Based on the results, the temperature dependence of the magnetothermal expansion is discussed in section 3, followed by concise summaries in section 4.

2. Magnetic properties of quasi 1-D itinerant magnets near the AF instability

Let us first discuss magnetic properties of itinerant quasi 1-D antiferromagnets based on the spin amplitude conservation by Takahashi [6]. It means that the local spin amplitude defined by

\[ \langle S_{\text{loc}}^2 \rangle = \frac{3}{N_0^2} \sum_{q} \int_{0}^{\infty} \frac{d\nu}{\pi} \text{coth}(\nu/2k_B T) \text{Im}\chi(Q + q, \nu), \]  

(2)

is almost conserved independent of the temperature and the presence of magnetic field. The total amplitude in (2) is divided into thermal and zero-point amplitudes defined by

\[ 3A_1(\kappa_s^2, T) \equiv \langle S_{\text{loc}}^2 \rangle_1 = \frac{6}{N_0^2} \sum_{q} \int_{0}^{\infty} \frac{d\nu}{\pi} \frac{1}{e^{\nu/k_B T} - 1} \text{Im}\chi(Q + q, \nu), \]

\[ 3A_2(\kappa_s^2) \equiv \langle S_{\text{loc}}^2 \rangle_2 = \frac{3}{N_0^2} \sum_{q} \int_{0}^{\infty} \frac{d\nu}{\pi} \text{Im}\chi(Q + q, \nu). \]

(3)

The imaginary part of the dynamical susceptibility for quasi 1-D itinerant magnets near the antiferromagnetic instability point is assumed to be given by the double Lorentzian form around the antiferromagnetic wave vector $Q$:

\[ \text{Im}\chi(Q + q, \nu) = \chi(Q + q, 0) \frac{\nu \Gamma_{Q+q}}{\nu^2 + \Gamma_{Q+q}^2}, \]

(4)

where $Q$ is of the order of the zone-boundary vector $q_B$ and the wave-vector dependence of the static magnetic susceptibility and the damping are given by

\[ \chi(Q + q, 0) = \frac{\chi(Q)\kappa_s^2}{\kappa_s^2 + q^2 + \varepsilon_a q^2}, \quad \Gamma_{Q+q} = \Gamma_Q(\kappa_s^2 + q_x^2 + \varepsilon_a q^2). \]

(5)

In (5), $\kappa_s^2 \propto \chi^{-1}(Q, 0)$ stands for the correlation wave-vector squared around the $Q$ vector, $q^2 = q_x^2 + q_y^2$, and a parameter $\varepsilon_a$ introduces an anisotropic wave vector dependence in the system, i.e., a quasi-one dimensionality. As measures of dispersions of the spectrum in $q, \nu$ spaces, two spectral widths are defined by

\[ T_0 = \frac{\Gamma_Q Q^2}{2\pi}, \quad \frac{N_0}{\chi(Q, 0)} = \frac{N_0}{\chi(Q, 0)} \left[ 1 + \frac{Q^2}{\kappa_s^2} \right] \approx \frac{N_0 Q^2}{\chi(Q, 0)\kappa_s^2} = 2T_A, \]

(6)
from differences of \(\chi(q,0)^{-1}\) and \(\Gamma q\) for \(q = (0, 0, Q)\) and \(0, 0, 0\) along \(q_z\)-axis. In the same way, the dispersions in the \(q_x, q_y\) plane are characterized by

\[
\frac{\Gamma_0 \varepsilon A Q^2}{2\pi} = \varepsilon A T_0, \quad \frac{N_0}{\chi(Q,0)} \simeq \frac{N_0 \varepsilon A Q^2}{\chi(Q,0) \kappa_s^2} = 2\varepsilon A T_A. \tag{7}
\]

from the differences between those for \(q = (\sqrt{\varepsilon A} Q, 0, 0)\) and \((0, 0, 0)\).

2.1. Temperature dependence of magnetic susceptibility

The total spin amplitude conservation can be explicitly written as

\[
A_t(\kappa^2_s, T) + A_z(\kappa^2_s) = \begin{cases} 
A_t(0, T_N) + A_z(0), & \text{for AF} \\
A_z(0) + [A_z(\kappa^2_s) - A_z(0)], & \text{for P} 
\end{cases} \tag{8}
\]

We define \(\kappa_{\delta 0}\) as \(\kappa_s\) in the ground state. The right hand side of (8) represents the amplitude for antiferromagnets at \(T = T_N\) or for paramagnets in the ground state, respectively. In the following, the above amplitudes, \(A_t(y,t)\) and \(A_z(y)\), are regarded as functions of reduced parameters \(y = \kappa^2_s/Q^2\) and \(t = T/T_0\) as defined below.

\[
A_t(y,t) = \frac{T_0 t}{T_A \varepsilon a} \int_0^1 \text{d}z [\phi(B(z,y)/t) - \phi(B(z,y + \varepsilon a)/t)] \\
\phi(x) = -(x - \frac{1}{2}) \ln x + x + \ln \Gamma(x) - \ln \sqrt{2\pi}, \quad B(z,y) = z^2 + y \\
A_z(y) \simeq A_z(0) - \frac{T_0}{T_A} c(\varepsilon a)y, \quad c(\varepsilon) = \frac{1}{\varepsilon} \log(1 + \varepsilon) + \frac{2}{\sqrt{\varepsilon}} \tan^{-1}\left(\frac{1}{\sqrt{\varepsilon}}\right)
\]

After substitution of the \(y\)-dependence of \(A_z(y)\) for \(y \ll 1\), both equations in (8) can be written in the same form:

\[
y = y_0 + d(\varepsilon a) t \int_0^1 \text{d}z [\phi(B(z,y)/t) - \phi(B(z,y + \varepsilon a)/t)], \quad d(\varepsilon) = \frac{1}{\varepsilon c(\varepsilon)}. \tag{9}
\]

The first constant \(y_0\) in the right hand side is the ground state value of \(y\) for P and is positive. It is negative for AF and is given by the thermal amplitude at \(T = T_N\):

\[
y_0 = -d(\varepsilon a) t_N \int_0^1 \text{d}z [\phi(B(z,0)/t_N) - \phi(B(z,\varepsilon a)/t_N)], \tag{10}
\]

where \(t_N = T_N/T_0\).

From the amplitude conservation (8) in the ground state, we can find the magnetic isotherm there under the presence of the staggered external magnetic field, giving \(\sigma_s^2 = 20A_t(0, T_N)\). It follows that the relation,

\[
\sigma_s^2 = 20A_t(0, T_N) = \frac{20 T_0 t_N}{T_A \varepsilon a} \int_0^1 \text{d}z [\phi(B(z,0)/t_N) - \phi(B(z,\varepsilon a)/t_N)], \tag{11}
\]

between the spontaneous moment \(\sigma_s\) and \(t_N\) is satisfied. In the 1-D limit, it is given by

\[
\sigma_s^2 = 20A_t(0, T_N) \simeq \frac{20 T_0 \pi t_N}{T_A \sqrt{\varepsilon a}}, \quad (\varepsilon a \rightarrow 0). \tag{12}
\]
Figure 1. The staggered spontaneous moment squared $\sigma_s^2$ versus $t_N$ for $\varepsilon_a = 0.05$, 0.1, 0.2, 0.5, and 1.0 from the top.

Figure 2. The enhancement of the thermal amplitude at $t_N$ in the 1-D limit.

The thermal amplitude is enhanced proportional to $\varepsilon_a^{-1/2}$ because of the development of the critical fluctuations by lowering the spacial dimensionality of the system. We show in Figure 1, the $t_N$-dependence of the thermal amplitude in the right hand side of (11) at $t = t_N$. The $t^{3/2}$-linear behaviors for 3-D antiferromagnets observed for larger $\varepsilon_a$ are found to crossover to the $t_N$-linear dependence in the vanishing $\varepsilon_a$ limit. In Figure 2, the thermal amplitude is plotted against $\varepsilon_a^{-1/2}$ in agreement with (12).

For paramagnets near the instability, the temperature dependence of the inverse of the reduced staggered susceptibility $y$ is numerically evaluated by solving (9). In the low temperature limit, the following $T^2$ dependence is obtained analytically [7]:

$$y \approx y_0 + \frac{1}{24} d(\varepsilon_a) t^2 \left[ \frac{1}{\sqrt{y}} \tan^{-1} \frac{1}{\sqrt{y + \varepsilon_a}} - \frac{1}{\sqrt{y + \varepsilon_a}} \tan^{-1} \frac{1}{\sqrt{y + \varepsilon_a}} \right],$$

$$\approx y_0 + \frac{\pi}{48} d(\varepsilon_a) t^2 \left[ \frac{1}{\sqrt{y_0}} - \frac{1}{\sqrt{y_0 + \varepsilon_a}} \right] \approx y_0 + \frac{1}{48\sqrt{\varepsilon_a}} t^2 \left\{ y_0^{-1/2}, \quad (y_0 \ll \varepsilon_a) \\
\varepsilon_a y_0^{-3/2}/2, \quad (y_0 \gg \varepsilon_a) \right. \right.$$  

(13)

for $y, \varepsilon_a \ll 1$ by using $d(\varepsilon) \approx \varepsilon^{-1/2}/\pi$ and the asymptotic behavior of $\phi(x)$,

$$\phi(x) \approx \frac{1}{12x}, \quad \text{for } x \gg 1.$$  

(14)

The $T^2$-linear coefficient shows crossover behavior from $(\varepsilon_a y_0)^{-1/2}$ for $y_0 \ll \varepsilon_a$ to $y_0^{-3/2}$ for $\varepsilon_a \ll y_0$ depending on the relative magnitude of $y_0$ and $\varepsilon_a$.

For antiferromagnets, the thermal amplitude near the critical point ($T_N \lesssim T$) can be written in the form,

$$\langle S^2 \rangle_t (y, t) \approx \langle S^2 \rangle_t (0, t) - \frac{3\pi T_N}{2T_A \varepsilon_a} \sqrt{y}, \quad \text{for } y < \varepsilon_a.$$  

(15)

Since the zero-point amplitude with $y$-linear dependence can be neglected around $T_N$, the temperature dependence of $y$ can be obtained by

$$\langle S^2 \rangle_t (0, t) - \frac{3\pi T_N}{2T_A \varepsilon_a} \sqrt{y} = \langle S^2 \rangle_t (0, t_N), \quad y = \left( \frac{2T_A \varepsilon_a}{3\pi T_N} \right)^2 \left[ \langle S^2 \rangle_t (0, t) - \langle S^2 \rangle_t (0, t_N) \right]^2.$$  

(16)
It means that \( y \) is proportional to \((T - T_N)^2\), and its coefficient has a \( \varepsilon_a \)-linear dependence because of the \( \varepsilon_a^{-1/2} \) dependence of the thermal amplitude.

We show in Figure 3 the temperature dependence of \( y \) for P at low temperatures for various values of \( \varepsilon_a \) from 0.05 to 1. The slope decreases with increasing \( \varepsilon_a \), for \( y_0 < \varepsilon_a \) is satisfied. In Figure 4, the temperature dependence of \( y \) for AF is shown as a function of \((t - t_N)^2\). In contrast to Figure 3, the slope increases with increasing \( \varepsilon_a \) because of the \( \varepsilon_a \)-linear slope for AF.

![Figure 3](image-url)  
**Figure 3.** Temperature dependence of \( y \) for P with \( y_0 = 0.01 \) and \( T_0/T_A = 0.1 \) as a function of \( t^2 \). Values of \( \varepsilon_a \) are shown in the key.

![Figure 4](image-url)  
**Figure 4.** The temperature dependence of \( y(t) \) for AF with \( t_N = 0.01 \).

3. Temperature dependence of thermal expansions

We employ the following free energy [3] consistent with the spin amplitude conservation to discuss the temperature dependence of thermal expansions, given for paramagnetic cases by

\[
F_m(y, T, V) = \frac{3}{\pi} \sum_q \int_0^\infty \mathrm{d}\nu \left[ \frac{\nu}{2} + T \ln(1 - e^{-\nu/T}) \right] \frac{1}{\nu^2 + \Gamma_{Q+q}^2 + \Gamma_{Q+q}^N} - N_0 T_A y \left\langle S_{loc}^2 \right\rangle .
\]

Because of the second term in the right hand side, its stability condition to the variation of \( y \) gives the amplitude conservation (2). The thermal expansion can be derived by introducing the volume dependence of parameters in the free energy. According to Grüneisen’s treatment of lattice vibrations, let us introduce the parameters,

\[
\gamma_0 = -\frac{\mathrm{d}\ln T_0}{\mathrm{d}\omega}, \quad \gamma_A = -\frac{\mathrm{d}\ln T_A}{\mathrm{d}\omega},
\]

that characterize the volume dependence of spectral parameters, \( T_0 \) and \( T_A \) [3]. The volume strain is defined by \( \omega = \delta V/V \). The thermal volume expansion is then derived from the explicit \( \omega \)-derivative of \( F_m \) under the stability condition of \( y \). The result consists of two terms from different origins. We are particularly concerned in this study with the term from the thermal spin fluctuations for paramagnets near the antiferromagnetic instability given by

\[
\omega_t(t) = 3\rho K \gamma_0 T_0 t \int_0^1 \mathrm{d}z \int_0^1 \mathrm{d}x \ u[\ln u - 1/2u - \psi(u)], \quad u = (y + z^2 + \varepsilon_a x)/t,
\]

where \( x \) is a variable related to the temperature and \( z \) is a variable related to the spin fluctuations.


where $\psi(u)$ is the digamma function.

At low temperatures, the temperature dependence of the thermal expansion $\omega_l(t)$ can be analytically obtained, showing the following $t^2$-linear dependence:

$$\omega_l(t) \simeq \frac{\rho K T_0 \gamma_0 t^2}{4 \varepsilon_a} \left( \ln \frac{1 + y_0 + \varepsilon_a}{1 + y_0} + 2 \sqrt{y_0 + \varepsilon_a} \tan^{-1} \frac{1}{\sqrt{y_0 + \varepsilon_a}} - 2 \sqrt{y_0} \tan^{-1} \frac{1}{\sqrt{y_0}} \right),$$

for $y_0, \varepsilon_a \ll 1$ by using the asymptotic expansion of the digamma function:

$$\ln u - \frac{1}{2u} - \psi(u) \simeq \frac{1}{12u^2}, \quad \text{for } u \gg 1.$$

The reciprocal magnetic susceptibility at $T = 0$ is denoted by $y_0$ as before. Its $t^2$-linear coefficient is therefore enhanced proportional to $\varepsilon_a^{-1/2}$ for $y_0 \ll \varepsilon_a$ and $y_0^{-1/2}$ for $\varepsilon_a \ll y_0$.

Figure 5 shows the temperature dependence of the thermal expansion $\omega_l(t)/3\rho K\gamma_0$ against $t^2$ for $y_0 = 0.01$ and various values of $\varepsilon_a$. Since $y_0 \ll \varepsilon_a$ is satisfied, slopes become steeper as $\varepsilon_a$ decreases. In Figure 6, the results with fixed $\varepsilon_a = 0.1$ are shown for $y_0 = 0.01, 0.05, \text{ and } 0.1$. Slopes of curves also become steeper as decreasing $y_0$ in agreement with the second line of (20).

![Figure 5](image_url) ![Figure 6](image_url)

**Figure 5.** The thermal expansion as a function of $t^2$ for fixed $y_0 = 0.01$ and $\varepsilon_a$ from 0.05 to 1.0 from the top.  
**Figure 6.** The $t^2$ linear behavior of $\omega_l(t)$ for $y_0 = 0.01, 0.05, \text{ and } 0.1$ with fixed $\varepsilon_a = 0.1$.

4. Summary

We have studied magnetic properties and thermal expansion of the quasi one-dimensional itinerant para- and antiferromagnets close to the magnetic instability point. By introducing the anisotropic parameter that controls the relative spectral dispersions along $q_z$-axis and in $q_x, q_y$-plane, we can continuously change the spacial dimension from 3-D to 1-D. We have found that magnetic properties especially around the critical point are sensitive to the effect of the change of spacial dimension as summarized below.

It is well-known that the relation, $\sigma^2 \propto t^3 \propto N^{-1}$, is satisfied for 3-D weak itinerant electron antiferromagnets. We found that it changes into the relation $\sigma^2 \propto t_N$ in the 1-D limit. The inverse staggered magnetic susceptibility shows $(t - t_N)^2$-linear dependence around the critical
point because of the critical spin fluctuations. The critical region of this behavior is found to increase by lowering the space dimension, for the coefficient of this term decreases towards this limit. The $t^2$-linear coefficients of the magneto-thermal expansion also show different enhancement around the magnetic instability point depending on the relative magnitude of $y_0$ and $\varepsilon_a$. Although it is difficult to find one-dimensional antiferromagnets, quasi one-dimensional compounds are in comparison easily available. The present results seem to suggest that quasi-low dimensional itinerant magnets in the extreme anisotropic limit are very interesting in our understanding the quantum critical behaviors from the observed crossovers by changing external parameters, i.e., the temperature, the magnetic field, the volume of crystals, and so on.

Acknowledgments
The authors thank the Yukawa Institute for Theoretical Physics at Kyoto University, where this work was initiated during the YITP-W-10-12 on “International and Interdisciplinary Workshop on Novel Phenomena in Integrated Complex Sciences: from Non-living to Living Systems”. This workshop was supported in part by the Grant-in-Aid for the Global COE Program “The Next Generation of Physics, Spun from Universality and Emergence” from the Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japan. This work was supported by the Kinki University Technical College Research Fund and a JSPS Grant-in-Aid for Science Research (C) 21540341.

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