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Magneto-volume effects in weakly ferromagnetic metals

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Abstract. Magneto-volume effects of weakly ferromagnetic metals are discussed, based on the assumption that the local spin fluctuation amplitude is almost constant. Our result on the thermal expansion agrees with the result of Moriya and Usami. We derive the universal relation of the ratio of thermal expansion above the Curie temperature and the spontaneous magnetostriction. Based on a simple thermodynamic argument, we also derive a relation connecting the magneto-volume coupling coefficient with the pressure dependence of the Curie temperature, which is slightly different from the Stoner-Wohlfarth theory.

1. Introduction

The self-consistent renormalization (SCR) spin fluctuation theory has so far been successful in explaining and predicting a lot of magnetic and other physical properties of weakly ferromagnetic metals [1]. Magneto-volume effects, however, have been mostly analyzed based on the Stoner-Wohlfarth (SW) theory [2], because until recently these effects have not been fully treated based on the SCR theory. Moriya and Usami [3] pointed out the importance of the effects of thermal spin fluctuations in dealing with the thermal expansion of the itinerant electron magnets and predicted different behaviours as compared with those by the SW theory, showing clearly the necessity of treating magneto-volume effects in these materials based on the SCR theory. In particular they predicted that the spontaneous magnetostriction, ω_s , is smaller than the value of ω_{SW} derived by the SW theory by a factor of 2/5, i.e. $\omega_s = 2\omega_{SW}/5$. They also pointed out the existence of positive magnetostriction even above the Curie temperature, whereas SW theory predicts no magnetostriction there. These predictions have been supported by the subsequent magneto-volume measurements on MnSi [4].

Recently, we showed that magnetic properties of weak ferromagnets can be treated based on a somewhat different point of view, i.e. from the assumption that the local spin fluctuation amplitude including both thermal and zero-point spin fluctuations is almost constant [5]. Later experimental investigations [6, 7] seem to support the consequence of this assumption. At first sight, this assumption seems in contradiction with the existence of magneto-volume effects in these materials. The magneto-volume effect is usually explained in association with the temperature variation of the local spin fluctuation amplitude. Therefore the purpose of the present paper is to study whether or not the above assumption is compatible with the magneto-volume effects. In previous investigations, the effect of zero-point spin fluctuations has been implicitly assumed to be very small. Only the effects of uniform magnetization, or the effects of both uniform magnetization and thermal spin fluctuations, have been considered. Neutron scattering experiments on some weak ferro- and antiferromagnets [8, 9] demonstrated the existence of a sizable contribution from zero-point spin fluctuations in these materials in the low-energy region. Therefore we explicitly take into account the effect of zero-point spin fluctuations. We point out that the wave-vector dependence of the magneto-volume coupling constant is important in deriving the magneto-volume effect of itinerant electron magnets. Our present result on the thermal expansion agrees with the previous results of Moriya and Usami. However, we add a new feature that the spontaneous magnetostiction) is a universal quantity. We present here a unified treatment including both the thermal expansion and the forced magnetostriction throughout the temperature range from below T_c to the paramagnetic phase. With the use of a simple thermodynamic argument, we also derive a relation connecting the magneto-volume coupling coefficient with the pressure dependence of the Curie temperature.

The plan of this paper is as follows. In the following section the wave vector and frequency dependence of the magneto-volume coupling constant is discussed. Both the effects of thermal and zero-point spin fluctuations on the volume striction are investigated here. Based on these results, temperature and external field dependences of volume striction are discussed in section 3. In section 4, we derive the relation between the forced magnetostriction coefficient and the pressure dependence of the Curie temperature. In the final section, conclusions and some discussions are presented.

2. Magneto-volume coupling constant

Before going into detail, we briefly summarize our theoretical framework. We assume that the local spin fluctuation amplitude, i.e. the sum of the average squared amplitudes of thermal and zero-point spin fluctuations and the uniform magnetization M is almost constant:

$$\langle S^2 \rangle = \langle S^2 \rangle_{\rm th} + \langle S^2 \rangle_{\rm zp} + \sigma^2 / 4 \tag{1}$$

where $\sigma = (M/N_0\mu_B)$ is the magnetization per magnetic atom in units of μ_B and N_0 the number of magnetic atoms in the crystal. In weak ferromagnets, at least for MnSi, the dynamical spin susceptibility, $\chi(q,\nu)$, is well described by the following formula in the small (q,ν) -region [10]:

$$Im\chi(q,\nu) = \frac{\chi(0)}{1+q^2/\kappa^2} \frac{\nu\Gamma_q}{\nu^2 + \Gamma_q^2}$$

$$\Gamma_q = \Gamma_0 q(\kappa^2 + q^2)$$
(2)

where $\chi(0)$ is the static uniform susceptibility and κ^2 is given by $\kappa^2 = N_0/2\bar{A}\chi(0)$. The parameter \bar{A} represents a constant characterizing the dispersion of the static magnetic susceptibility in q-space, whereas Γ_0 a measure of the energy distribution of the spin fluctuation spectrum. With the use of (2) and the fluctuation-dissipation theorem, the amplitude of zero-point spin fluctuation can be expressed in the form [5]:

$$\langle S^2 \rangle_{\rm zp} = \langle S^2 \rangle_{\rm zp} (T_{\rm c}) - \frac{3T_0}{2T_A} (2y_{\rm t} + y_{\rm l}) \tag{3}$$

where T_0 and T_A are defined by

$$T_0 = \Gamma_0 q_{\rm B}^3 / 2\pi \qquad T_A = \bar{A} q_{\rm E}^2$$

with $q_{\rm B}$, the effective zone-boundary wave number, equal to $(6\pi^2 N_0/V)^{1/3}$. In (3) $y_{\rm t}$ and $y_{\rm l}$ represent the reduced reciprocal susceptibilities of transverse and longitudinal components explicitly given by

$$y_{\rm t} = \frac{1}{k_{\rm B}T_A}\frac{h}{\sigma} \qquad y_{\rm l} = \frac{1}{k_{\rm B}T_A}\frac{\partial h}{\partial \sigma}$$
 (4)

where h is the external field, $2\mu_{\rm B}H$. We have shown that the magnetic properties of weak ferromagnets can be derived from the above assumption in terms of the two parameters, T_0 and T_A [5].

Now let us discuss the magneto-volume coupling vertex function in some detail. We are especially concerned with the vertex function diagrammatically shown in figure 1, which has two external spin fluctuation lines and one phonon line. The free energy is expanded in terms of the wave vector and frequency-dependent spin fluctuation amplitudes by

$$F = \frac{V}{2\kappa}\omega^{2} + \frac{1}{2}\sum_{q,\nu}\chi^{-1}(q,\nu)S_{q,\nu} \cdot S_{-q,-\nu} + \cdots$$
(5)

where the first term represents the elastic energy of the crystal with the volume, V, and the compressibility, κ , and ω represents the volume striction, $\delta V/V$. The magneto-volume coupling constant is then given by

$$\frac{1}{N_0}\lambda(q,\nu) = -\frac{1}{2}\frac{\partial}{\partial\omega}\chi^{-1}(q,\nu)$$
(6)

and the volume magnetostriction by

$$\omega = \frac{\kappa}{VN_0} \sum_{q,\nu} \lambda(q,\nu) \langle \boldsymbol{S}_{q,\nu} \cdot \boldsymbol{S}_{-q,-\nu} \rangle = \omega_{\rm th} + \omega_{\rm zp} \tag{7}$$

where $\langle \cdots \rangle$ represents the thermal average.



Figure 1. The magneto-phonon coupling constant. The broken line and wavy lines represent phonon and spin fluctuation propagators, respectively.

As shown in (7) magneto-volume vertex function, $\lambda(q,\nu)$, in general has q,ν dependences. For the volume striction $\omega_{\rm th}$ from the uniform magnetization and the thermal spin fluctuations, we can replace $\lambda(q,\nu)$ by its static uniform value, λ_0 , since thermal fluctuation amplitudes are sharply peaked around q = 0 and $\nu = 0$. From (1) and (3) $\omega_{\rm th}$ is therefore given by

$$\omega_{\rm th} = \kappa \lambda_0 \frac{N_0}{V} (\langle S^2 \rangle_{\rm th} + \sigma^2 / 4)$$

= $\kappa \lambda_0 \frac{N_0}{V} \left(\langle S^2 \rangle - \langle S^2 \rangle_{\rm zp} (T_{\rm c}) + \frac{3T_0}{2T_A} (2y_{\rm t} + y_{\rm l}) \right).$ (8)

Because the q, ν -dependences of zero-point spin fluctuation amplitudes is, on the contrary, relatively weak, we need to explicitly take into account these dependences of the magneto-volume coupling constant. The main temperature and field dependences of the zero-point spin fluctuation amplitudes come through those of y. Therefore what we want to know is the y-dependence of the zero-point spin fluctuation part ω_{zp} in the right-hand side of (7). As far as the y-dependence is concerned, only the low-frequency part has a significant effect in the sum in (7). We suppose that the ν -dependence of $\lambda(q, \nu)$ is considered to be weak in the energy interval up to the energy of the order of $k_B T_0$, where Im $\chi(q, \nu)$ has a significant weight. The ν -dependence of $\lambda(q, \nu)$ will play minor role as will be seen below.

In the paramagnetic phase, volume striction, ω_{zp} , from zero-point spin fluctuations is given by

$$\omega_{zp} = \frac{3\kappa}{VN_0} \sum_q \int_0^\infty \frac{d\nu}{\pi} \lambda(q,\nu) \operatorname{Im}\chi(q,\nu)$$

= $\frac{3\kappa}{\pi V} \sum_q \frac{\Gamma_0}{2\bar{A}} \frac{q}{2} \{\lambda(q,\nu_c) \log(\nu_c^2 + \Gamma_q^2) - \lambda(q,0) \log \Gamma_q^2$
 $- \int_0^{\nu_c} d\nu \lambda'(q,\nu) \log(\nu^2 + \Gamma_q^2) \}$
= $\operatorname{constant} - \frac{3\kappa}{\pi V} \sum_q \frac{\Gamma_0}{2\bar{A}} q\lambda(q,0) \log \Gamma_q$ (9)

where ν_c is the cut-off energy which may depend on q. In the above equation, λ' represents the first derivative of λ with respect to ν . Because this quantity will be very small in the low-energy region of interest and $\Gamma_q/\nu_c \ll 1$, the y-dependence mainly comes from the second term of the last line in (9). By expanding the last term in y, the y-dependence of ω_{zp} is expressed in the form

$$\omega_{zp} = \text{constant} - d\kappa \lambda_0 \frac{N_0}{V} \frac{3T_0}{2T_A} 3y + \cdots$$

$$d = \frac{1}{\lambda_0} \int_0^1 dx^2 \,\lambda(q_B x, 0).$$
(10)

In the ordered state, it is easily seen that 3y in (10) is replaced by $2y_t + y_1$.

Adding the above two contributions, the volume striction from the magnetic origin is now given by

$$\omega = \frac{\kappa \lambda_0 N_0}{V} (1 - d) \frac{3T_0}{2T_A} (2y_t + y_l) = \rho \kappa C' (2y_t + y_l).$$
(11)

If there is no q-dependence of $\lambda(q, 0)$, it follows that we have no magneto-volume effects. We do not know what value d will take. Here we simply assume that (1 - d) is finite and has a value of order 1. Equation (11) is the central result of this paper which is valid throughout all the temperature range from below T_c to above T_c under the external field, h.

3. Thermal expansion and forced magnetostriction

The temperature and field dependences of volume striction are discussed in this section based on (11). According to this equation, temperature and external field dependence of the volume striction can be evaluated from those of the reciprocal transverse and longitudinal susceptibilities, y_t and y_l . Let us first consider the magneto-volume effects in the ground state. At T = 0 K, uniform magnetization obeys the following equation:

$$h = \frac{1}{8}\bar{F}_1\sigma(\sigma^2 - \sigma_s^2) \tag{12}$$

where σ_s is the saturation magnetization per magnetic atom at T = 0 K in units of $2\mu_B$, which is related to the Curie temperature T_c by

$$\frac{\sigma_{\rm s}^2}{4} = \frac{15T_0}{T_A} c\eta^4 \qquad \eta = (T_{\rm c}/T_0)^{1/3}.$$
(13)

The numerical constant c is given by $0.3353\cdots$, whereas \bar{F}_1 is the fourth-order expansion coefficient of the free energy with respect to the uniform magnetization. Equation (13) is experimentally very well confirmed [11,12]. We have derived in the previous paper [5] that \bar{F}_1 is given by

$$\bar{F}_1 = \frac{4}{15} \frac{k_{\rm B} T_A^2}{T_0}.$$
(14)

From the definitions (4) and (12) we can easily see that

$$k_{\rm B}T_A y_{\rm t} = \frac{1}{8}\bar{F}_1(\sigma^2 - \sigma_{\rm s}^2) \qquad k_{\rm B}T_A y_{\rm l} = \frac{1}{8}\bar{F}_1(3\sigma^2 - \sigma_{\rm s}^2).$$
(15)

Now from (11) we can immediately obtain the following expression of the volume striction in the ground state:

$$\omega = \rho \kappa C' \frac{\bar{F}_1}{8k_{\rm B}T_A} (5\sigma^2 - 3\sigma_{\rm s}^2) \tag{16}$$

and forced and spontaneous magnetostrictions, ω_h and ω_s , i.e.

$$\omega_h = \rho \kappa C_h \sigma^2 \qquad \omega_s = \rho \kappa C_s \sigma_s^2 \tag{17}$$



Figure 2. $\Delta l/l - M^4$ plot for MnSi at T = 29 K.

where

$$C_{h} = \frac{5\bar{F}_{1}}{8k_{\rm B}T_{A}}C' = \frac{\lambda_{0}N_{0}}{4\rho V}(1-d) \qquad C_{\rm s} = \frac{2}{5}C_{h}$$

Because in the SW theory, ω_s is given by $(\rho \kappa C_h) \sigma_s^2$, it follows that $\omega_s / \omega_{SW} = 2/5$, in agreement with the result of Moriya and Usami [3].

In order to discuss the temperature and field dependences of volume striction at an arbitrary temperature and field strength, we have to know these dependences of y_t and y_l . We have already given a prescription to calculate them. We do not repeat it here. We here only point out that from (11) volume magnetostriction can be expressed in the following general form:

$$\omega = \frac{8}{5\bar{F}_1} (\rho \kappa C_h) \left(\frac{\partial h}{\partial \sigma} + \frac{2h}{\sigma}\right) = \omega_s \chi^{zz}(0) \left(\chi^{zz}(T,H)^{-1} + 4\chi^{-+}(T,H)^{-1}\right)$$
(18)

with

$$\chi^{zz}(0) = 2N_0 / \bar{F}_1 \sigma_{\rm s}^2$$

where χ^{zz} and χ^{-+} are the longitudinal and transverse magnetic susceptibilities in units of $(2\mu_{\rm B})^2$ given by $N_0\partial\sigma/2\partial h$ and $N_0\sigma/h$, respectively. Because of the rotational symmetry, $\chi^{-+}(T)^{-1}$ always vanishes identically below T_c when h = 0. Therefore without an external field the volume striction ω is proportional to $\chi^{zz}(T)^{-1}$ below T_c . Equation (18) also shows that the simple $\omega \propto M^2$ relation is not in general correct. Especially at the critical point, we have previously shown that the M-H curve obeys the following relation [5]:

$$H \propto M^5$$
 (19)

when M is small. Therefore both y_t and y_l are proportional to M^4 . This means that the forced magnetostriction at $T = T_c$ behaves as

$$\omega_h \propto M^4.$$
 (20)

This behaviour is clearly seen in the forced magnetostriction experiment on MnSi [4]. As shown in figure 2, the linearity of $\Delta l/l-M^4$ plot is very good except for some deviations in the very small M region.

In the paramagnetic phase according to (18) there is the positive magnetostriction given by

$$\omega = 3\omega_{\rm s}\frac{\chi^{zz}(0)}{\chi(T)} = \frac{3}{4c\eta^4}\omega_{\rm s}y\tag{21}$$

where we have used the relation $\chi^{zz}(T) = \chi^{-+}(T)/2 = \chi(T)$ at h = 0. We have shown that the (T/T_c) -dependence of y is solely determined by the value $\eta = (T_c/T_0)^{1/3}$ [5]. It then follows from (18) that the (T/T_c) -dependence of ω/ω_s is also determined by the value of η . In other words, the thermal expansion $\alpha(= d\omega/3dT)$ in the paramagnetic phase is given by

$$\frac{T_{\rm c}\alpha}{\omega_{\rm s}} = \frac{1}{4c\eta^4} \frac{\mathrm{d}y}{\mathrm{d}t} \qquad t = T/T_{\rm c} \tag{22}$$

where the t-dependence of the right-hand side depends on a single parameter η . We have numerically calculated (22) for several values of T_c/T_0 . The results are shown in figure 3. As shown in this figure, the value of $T_c\alpha/\omega_s$ has an almost constant value above T_c , with a larger value for smaller T_c/T_0 values. In figure 4 we have plotted this value estimated at t = 5 against T_c/T_0 . The magnetic contribution of the thermal expansion above T_c has been experimentally estimated for MnSi and Ni₃Al. The values of $T_c\alpha/\omega_s$ above T_c are given by 0.27 for MnSi [4] and 1.1 for Ni₃Al [13]. On the other hand, from the estimated values of T_c/T_0 , i.e. 0.13(MnSi) and 0.012(Ni₃Al), the values of $T_c\alpha/\omega_s$ are predicted to be 0.3 for MnSi and 0.6 for Ni₃Al from figure 4, showing fair agreement with the above observed values. A recent magneto-volume experiment on the (Fe_{1-x}Co_x)Si alloy system also seems to support our predictions [7].



Figure 3. Temperature dependence of the thermal expansion coefficient.

If we assume that χ obeys the Curie-Weiss law

$$\chi(T) = \frac{N_0 p_{\rm eff}^2}{12k_{\rm B}(T-T_{\rm c})}$$

the thermal expansion, α , above $T_{\rm c}$ from the magnetic origin, is also expressed in the form

$$\alpha = k_{\rm B}\omega_{\rm s} \frac{12}{p_{\rm eff}^2} \frac{\chi^{zz}(0)}{N_0} = \omega_{\rm s} \frac{24}{p_{\rm eff}^2} \frac{k_{\rm B}}{\bar{F}_1 \sigma_{\rm s}^2}.$$
(23)



Figure 4. $T_c \alpha / \omega_s - T_c / T_0$ plot.

With this formula we can estimate the thermal expansion of the magnetic origin in the paramagnetic phase. Equation (23) formally agrees with the result of Moriya and Usami. The difference is that the ratio of $T_c \alpha / \omega_s$ is universal in the present treatment and is uniquely determined by the value of T_c / T_0 . In principle this ratio may take any value. Observed ratios, however, seem to fall in a restricted range.

4. Forced magnetostriction coefficient

Let us begin with the following thermodynamic argument. The free energy deviation against variations of pressure p and an external field h is given by

$$\mathrm{d}F = V\omega\mathrm{d}p - \frac{1}{2}N_0\sigma\,\mathrm{d}h.\tag{24}$$

From (24) the following Maxwell relation is derived:

$$\left(\frac{\partial\omega}{\partial h}\right)_{p} = -\frac{N_{0}}{2V} \left(\frac{\partial\sigma}{\partial p}\right)_{h} = -\frac{N_{0}}{2V} \sigma \left(\frac{\partial\ln\sigma}{\partial p}\right)_{h}.$$
(25)

On the other hand, if we define forced magnetostriction coefficient by the following equation in the ground state

$$\omega = \omega_0 + \rho \kappa C_h \sigma^2 \tag{26}$$

the forced magnetostriction is also expressed as follows:

$$\frac{\partial \omega}{\partial h} = 2(\rho \kappa C_h) \sigma \frac{\partial \sigma}{\partial h} = 4(\rho \kappa C_h) \sigma \frac{\chi^{zz}(0)}{N_0}.$$
(27)

By equating (25) with (27), we obtain the following relation:

$$(\rho\kappa C_h) = -\frac{N_0}{8V} \frac{N_0}{\chi^{zz}(0)} \left(\frac{\partial \ln \sigma}{\partial p}\right)_h$$
(28)

which is valid regardless of our choice of approximation.

	σ	d <i>T</i> c/dp (K/kbar)	$ar{F}_1/k_{ m B}$ (K)	$(ho\kappa C_h)^{ m cal}$ $(g/ m emu)^2$	$(ho\kappa C_h)^{ m obs}$ $(g/ m emu)^2$
Ni ₃ Al	0.075	-0.50	1.33×10^{5}	0.50×10^{-6}	0.64×10^{-6}
ZrZn ₂	0.12	-1.8	1.54×10^{4}	4.22×10^{-6}	5.03 x 10 ⁻⁶
MnSi	0.4	-1.17	0.82×10^4	2.75×10^{-6}	1.03×10^{-6}
Sc_3In	0.045	0.19	1.63×10^{5}	-1.3×10^{-6}	-1.6×10^{-6}

Table 1. Observed and calculated forced magneto-striction coefficients.

Our next step is to relate (28) with the pressure dependence of T_c . If we assume that the pressure dependence of the right-hand side of (13) is mainly given by that of T_c , we immediately obtain the following relation:

$$\left(\frac{\partial \ln \sigma}{\partial p}\right)_{h} = \frac{2}{3} \left(\frac{\partial \ln T_{c}}{\partial p}\right)_{h}.$$
(29)

By substituting (29) into (28), we finally obtain the following relation:

$$(\rho\kappa C_h) = -\frac{N_0}{12V} \frac{N_0}{\chi^{zz}} \left(\frac{\partial \ln T_c}{\partial p}\right)_h = -\frac{N_0}{24V} \sigma_s^2 \bar{F}_1 \left(\frac{\partial \ln T_c}{\partial p}\right)_h.$$
 (30)

It should be noted that (30) is derived from the Maxwell relation and the magnetic equation of state (15) in the ground state and is free from the relation (14). We also note that in the SW theory, because $\sigma_s^2 \propto T_c^2$, a value of $(\rho \kappa C_h)$ is obtained which is 1.5 times greater.

In (29) we have neglected the volume dependence of T_0 and T_A . If we expand the non-interacting dynamical susceptibility in the small q, ν - region as follows

$$\chi_0(q,\nu)/\chi_0(0,0) = 1 - Aq^2 + \dots + iC\nu/q + \dots$$
 (31)

then the above parameters are explicitly given by

$$T_0 = \frac{Aq_{\rm B}^3}{2\pi C} \qquad T_A = \frac{N_0}{\rho(\varepsilon_{\rm f})} Aq_{\rm B}^2$$
(32)

with $\rho(\varepsilon_{\rm f})$, the density of states at the Fermi level, $\varepsilon_{\rm f}$. In order to get the order of magnitude estimate of the volume dependence of parameters T_0 and T_A , let us assume a free electron-gas like dispersion relation for the band electrons in the crystal. Then the following expressions are obtained:

$$k_{\rm B}T_0 = \frac{24}{\pi^2} (q_{\rm B}/k_{\rm f})^3 \varepsilon_{\rm f} T_0/T_A = 2(12/\pi)^2 (q_{\rm B}/k_{\rm f}) (\rho(\varepsilon_{\rm f})\varepsilon_{\rm f}/N_0).$$
(33)

If we further assume Heine's [14] uniform $V^{5/3}$ -dependence of the d-band width, we obtain the value, -5/3, for $\partial \ln \varepsilon_{\rm f}/\partial \omega$ and $\partial \ln T_0/\partial \omega$. On the other hand, the value $\rho(\varepsilon_{\rm f})\varepsilon_{\rm f}$ will show little volume dependence. Because the observed volume derivative for $\ln T_{\rm c}$ has a value of around 30 for Ni₃Al, the pressure dependence of these parameters, T_0 and T_A , are an order of magnitude smaller than $dT_{\rm c}/dp$.

In order to assess the validity of (30) we have estimated the left-hand side of (30)using the observed parameter values of σ_s , $\partial T_c/\partial p$, and \bar{F}_1 , for weak ferromagnets, Ni₃Al [13, 15-17], ZrZn₂ [18-22], MnSi [4, 23, 24], and Sc₃In [25, 26]. We used the values of \overline{F}_1 from the observed Arrott plots at low temperature rather than (14). Indirect measurements of T_0 and T_A through NMR relaxation measurements and neutron scattering experiments may have some error. The results are shown in table 1 by $(\rho \kappa C_h)^{cal}$ together with the observed values for the forced magnetostriction coefficients. Agreements are fairly good. Although we cannot decisively conclude the validity of our formula (30) from this table, the analysis seems to slightly favour the SCR theory. A recent measurement on $(Fe_{1-x}Co_x)Si$ system also supports (28) [7]. Another test is to directly check (29) by measurements of the pressure derivatives of $\ln \sigma$ and $\ln T_c$. We also show in table 2 the observed values for these quantities. As seen in this table, observations on the Ni₃Al system seem to strongly support our (29), although for other materials the agreements are relatively poor because of several experimental uncertainties. For example, if we use (28) to evaluate $(\rho \kappa C_h)$ for MnSi, the value 1.22×10^{-6} (g/emu)² is obtained, in close agreement with experiment. Because (28) is the direct consequence of the Maxwell relation, the discrepancy in table 1 for MnSi is thus ascribed to the observed value of $\partial \ln T_c / \partial p$.

Table 2. Observed pressure dependences of the saturation moment σ and $T_{\rm c}$.

	$\frac{-\mathrm{d}\ln\sigma/\mathrm{d}p}{(10^{-3} \mathrm{kbar}^{-1})}$	$-1.5 d \ln \sigma / d p$ (10 ⁻³ kbar ⁻¹)	$-d \ln T_c/dp$ (10 ⁻³ kbar ⁻¹)
Ni ₇₆ Al ₂₄	6.27	9.41	5.0
Ni75.5 Al24.5	5.55	8.33	7.12
Ni75 Al25	8.69	13.0	11.6
Ni74.8 Al25.2	13.6	20.4	19.3
ZrZn _{1.9}	44	66	47
MnSi	11.5	17.3	39

5. Conclusions and discussions

In the present paper we have discussed the magneto-volume effects arising from both the thermal and zero-point spin fluctuations. The essence of this paper depends on the assumption of the q-dependence of the magneto-volume coupling constant. This may seem to contradict to our conventional view. Note, however, that in MnSi the observed q, ν -integrated spin fluctuation amplitude shows very weak temperature dependence [8]. Nevertheless, this material shows large magneto-volume effects. The same behaviour was also observed in (YSc)Mn₂ [9]. Because the energy integration is not wide enough and the resolution is not so good, we cannot definitely conclude the weak temperature dependence of the local spin fluctuation amplitudes from these experiments. But we do not expect that the amplitude will show considerable temperature dependence when higher energy excitations are included. Then, if we assume a uniform magnetostriction coupling, we have to assume a quite large magneto-volume coupling constant in order to explain observed magneto-volume effects. Therefore this problem is still open to further studies. We derived the general formula (18) of the volume striction, which is valid throughout the temperature range from below T_c to above T_c at an arbitrary field strength. Here, both the thermal expansion and forced magnetostriction are treated on an equal footing. We showed that the forced magnetostriction is not proportional to σ^2 in general, but to the reciprocal susceptibility. As an application we showed that $\Delta l/l \propto M^4$ when $T \sim T_c$. This will serve as a test of our predictions. The forced magnetostriction measurement on MnSi near the Curie temperature seems to support our relation. As far as the thermal expansion is concerned, our results formally agree with those of Moriya and Usami. A new feature derived here is that the ratio $T_c \alpha/\omega_s$ is the universal quantity, solely determined by T_c/T_0 .

Another point of this paper is concerned with the relation between the magnetovolume coupling coefficient and the pressure dependence of the Curie temperature. It is shown there exists a slight difference in this relation between the SW and the SCR theories. It seems that present experimental data slightly favours the SCR theory. We hope more detailed and accurate experimental data on various weak ferromagnets will be compiled and analyzed in order to clarify this point.

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