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Magnetic properties of Co_2TiGa compound

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Abstract

The precise magnetization measurement under ambient pressure has been made for Co_2TiGa using a SQUID magnetometer. The spontaneous magnetization $\sigma_s(T)$ at ambient pressure is expressed empirically as a function of temperature as $\sigma_s(T)^2=\sigma_s(0)^2(1-T^2/T_C^2)$, where $\sigma_s(0)$ and T_C are 19.5 emu/g, 128 K, respectively. The pressure derivative of T_C has been obtained from the results of temperature dependence of initial permeability under pressure up to 11 kbar. The value of $\partial T_C/\partial P$ is -1.27 K/kbar. The results are discussed on the basis of the spin fluctuation theory for itinerant electron magnetism.

Keywords: Co_2TiGa; Magnetization measurement; Pressure effect; Spin fluctuation

1. Introduction

The pressure effect on the Curie temperature T_C was intensively studied for the manganese Heusler alloys Cu_2MnAl, Ni_2MnZ (Z=Al, Ga, Sn and Sb), Au_2MnAl and Pd_2MnZ (Z=Sn and Sb), in which Mn atoms have definite localized moments of about 4 μ_B [1–8]. For all alloys, the Curie temperature increases linearly with increasing pressure. On the basis of the results, the interatomic distance dependence of the exchange interaction was discussed for a number of manganese Heusler alloys. However, there has been only a little amount of information about the magnetovolume effect for cobalt Heusler alloys, which are typical itinerant electron ferromagnets. Previously, Kanomata et al. obtained the pressure derivative of the Curie temperature $\partial T_C/\partial P$ for cobalt Heusler alloys Co_2TiAl and Co_2TiGa from the results of temperature dependence of initial permeability under pressures up to 5 kbar [9]. The values of $\partial T_C/\partial P$ were reported to be $+0.6$ K/kbar for Co_2TiAl and -1.3 K/kbar for Co_2TiGa. Then, DiMasi et al. measured the electrical resistivity at temperature between 1.2 and 300 K and pressures up to 12 kbar to determine the pressure derivative of the Curie temperature for Co_2TiAl [10]. They reported the value of -0.7 K/kbar as $\partial T_C/\partial P$ and emphasized the importance of the contribution of spin fluctuation in understanding the magnetovolume effect of cobalt Heusler alloys.

In this paper, the precise magnetic properties of Co_2TiGa are examined. And then, the pressure dependence of the Curie temperature of Co_2TiGa is reported. The experimental results are discussed using a spin fluctuation theory developed by Takahashi [11].

2. Experimental

The ordered alloy Co_2TiGa was prepared by repeated melting of appropriately composed mixtures of 99.9% pure Co, 99.9% pure Ti and 99.9999% pure Ga in an argon arc furnace. Since the weight loss after melting was negligible, the nominal composition was accepted as being accurate. To get the homogenized sample, the reaction product was sealed in evacuated silica tube, heated at 850°C for 7 days and then quenched in water. X-ray diffraction spectrum was taken with CuKα radiation on powder sample. All diffraction lines were indexed with the cubic structure. The lattice parameter was found to be $a=5.857$ Å. It's value is in good agreement with that reported by Webster and Ziebeck [12]. The degree of atomic ordering for the sample prepared as above can be estimated by comparing the experimental values of relative intensities of even ($h+k+l=4n+2$) and odd (h,k,l=all odd) superstructure lines of $L2_1$ structure with those expected from ideal atomic ordering. The observed values of $|F(2\ 0\ 0)|^2/|F(2\ 2\ 0)|^2$ and $|F(1\ 1\ 1)|^2/|F(2\ 2\ 0)|^2$ indicate that the sample in this study has a fully ordered Heusler structure, where $F(2\ 2\ 0)$

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means the structure amplitude of the fundamental line (2 2 0).

Magnetization of Co_2TiGa at ambient pressure was measured in magnetic fields up to 50 kOe at temperature from 5 to 350 K using a commercial superconducting quantum interference device (SQUID) magnetometer.

Hydrostatic pressure was applied to the sample in a teflon pressure sample cell filled with a silicon oil by using a piston-cylinder type device. The Curie temperature at high pressure was determined by an ac transformer method, where the primary and secondary coils were wound on the sample. An ac current of a constant amplitude flowed in the primary coil and the second voltage, which is directly proportional to initial permeability, was recorded as a function of temperature at various pressures. The pressure was calibrated by using the Hg solid–liquid transition temperature.

3. Results and discussion

Fig. 1a and b show the magnetization curves of Co_2TiGa with $5 \leq T \leq 160$ K and $180 \leq T \leq 350$ K in magnetic fields up to 50 kOe, respectively. As seen in Fig. 1a, the magnetization curves are characteristic for ferromagnets. The magnetization σ at 5 K is saturated in the magnetic field of about 5 kOe, indicating that the magnetic crystalline anisotropy energy of Co_2TiGa is small. The magnetization above $T \geq 180$ K increases linearly with increasing applied field. We determined the temperature dependence of the susceptibility χ for Co_2TiGa from the slope of the magnetization curves in Fig. 1b. The result is shown in Fig. 2. The observed $1/\chi$ vs. T curve can be well expressed by the Curie–Weiss law. From this result, the paramagnetic Curie temperature θ_p and the effective paramagnetic moment p_{eff} were determined to be 137 K and 2.13 μ_B, respectively. The paramagnetic moment p_c is obtained as $p_c = 1.35$ μ_B, where $p_{eff}^2 = p_c(p_c + 2)$. Isothermal $\sigma(H,T)^2$ vs. $H/\sigma(H,T)$ plot, i.e. the so-called Arrott plots, is a useful way to characterize the magnetic properties of magnetic material. The result of the Arrott plot is shown in Fig. 3 for Co_2TiGa. As seen in the figure, the Arrott plots give series of almost parallel straight lines over a wide temperature range below and above T_C, showing that the sample is good homogeneous ferromagnet. Spontaneous magnetization $\sigma_s(T)$ was determined by the linear extrapolation to $H/\sigma(H,T) = 0$ of the $\sigma(H,T)^2$ vs. $H/\sigma(H,T)$ curves. The magnetic moment at 5 K for Co_2TiGa is found to be 0.82 μ_B, in good agreement with the value reported by Ziebeck and Webster [13]. The value of the Curie temperature was defined as the temperature at which an extrapolated curve of the Arrott plots goes through the origin. The Curie temperature of Co_2TiGa was determined to be $T_C = 128$ K. Fig. 4 shows the temperature dependence of the square of the spontaneous magnetization, $\sigma_s(T)^2$, against T^2. As seen in the figure, $\sigma_s(T)^2$ of Co_2TiGa

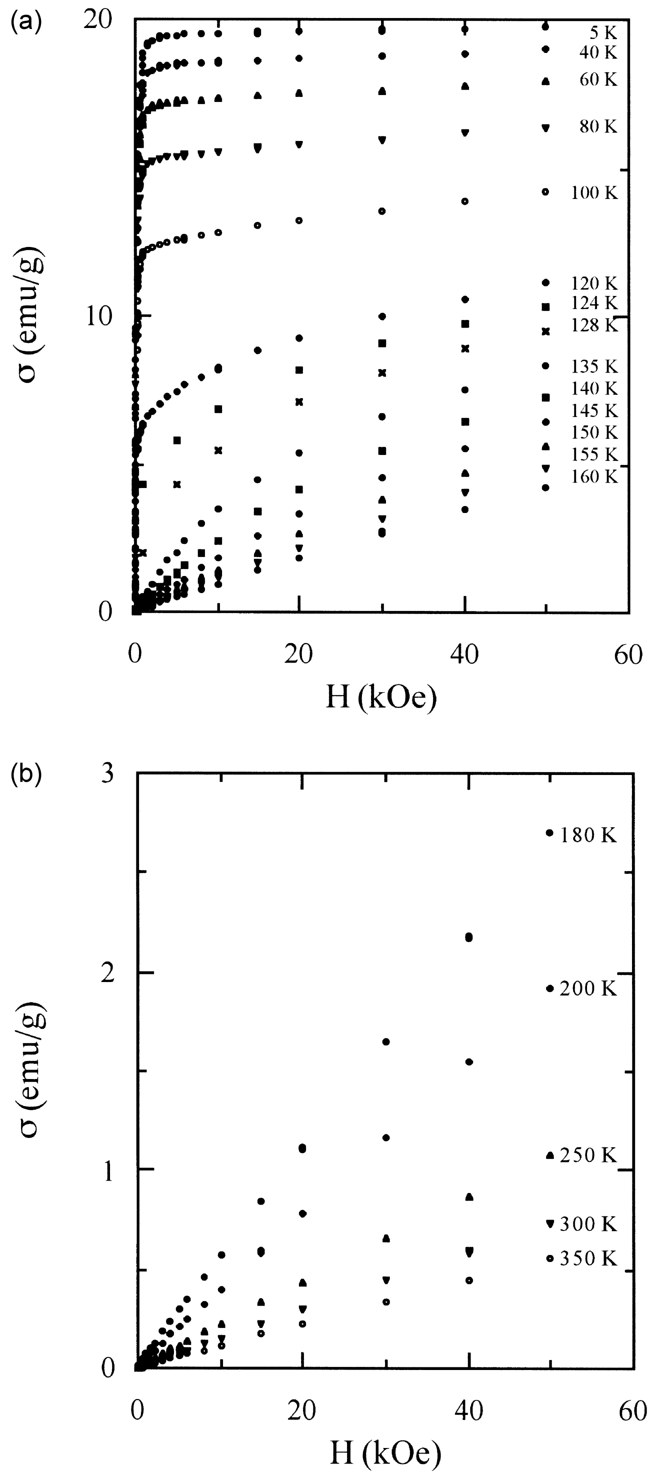

Fig. 1. (a) Magnetization curves of Co_2TiGa at various temperatures ($5 \leq T \leq 160$ K). (b) Magnetization curves of Co_2TiGa at various temperatures ($180 \leq T \leq 350$ K).

shows a T^2 dependence in a wide temperature range below T_C. That is, $\sigma_s(T)$ is expressed empirically as a function of temperature as

$$\sigma_s(T)^2 = \sigma_s(0)^2(1 - T^2/T_C'^2) \quad (1)$$

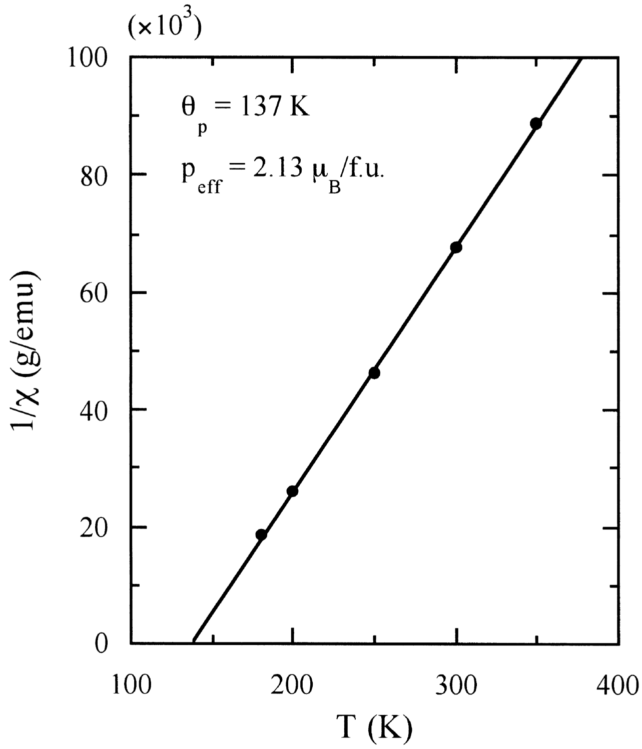

Fig. 2. Temperature dependence of the inverse susceptibility $1/\chi$ for Co_2TiGa.

The constant $T_C' = 128$ K thus determined is in very good agreement with the Curie temperature of Co_2TiGa.

Takahashi developed the spin fluctuation theory for the weak itinerant electron ferromagnets [11]. According to his theory, the magnetization of a weak itinerant electron

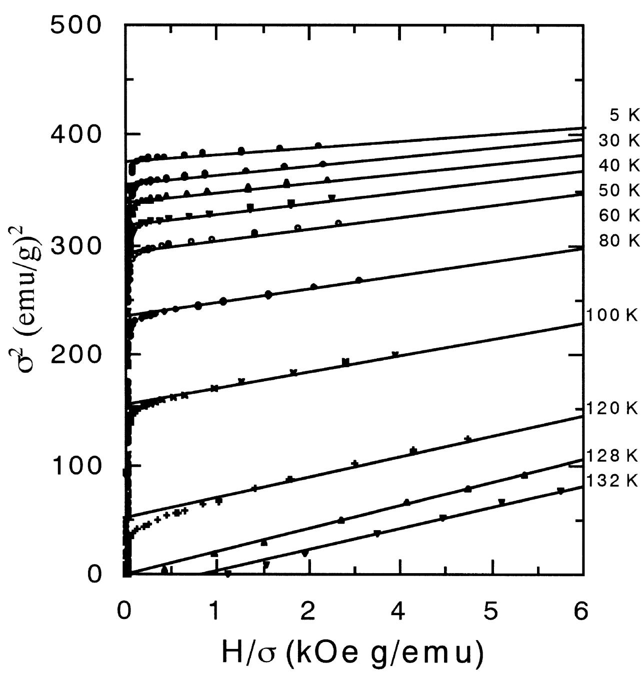

Fig. 3. $\sigma(T,H)^2$ vs. $H/\sigma(T,H)$ plots for Co_2TiGa at various temperatures.

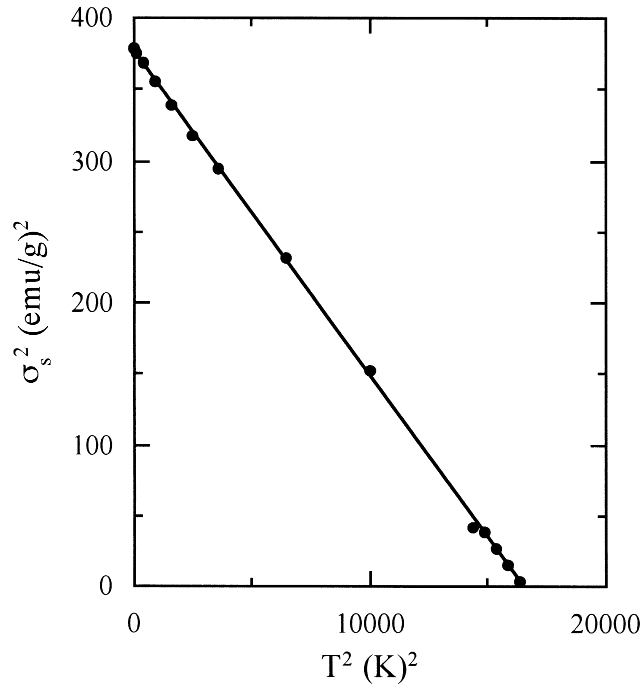

Fig. 4. Square of the spontaneous magnetization $\sigma_s(T)^2$ vs. T^2 plot for Co_2TiGa.

ferromagnet in the ground state is expressed by the following equation,

$$-2c\eta^4 k_B T_A\, p + \frac{4k_B T_A^2}{15T_0}\frac{p^3}{8} = h \tag{2}$$

with

$p = \sigma(H,T)/\mu_B N_0$ (here $T = 0$ K)

$h = 2\mu_B H$

$\eta^3 = T_C/T_0$

$c = 0.3353 \cdots$

where k_B is the Boltzmann constant, N_0 the number of magnetic sites. The parameters T_0 and T_A characterize the energy width of the dynamical spin fluctuation spectrum and the dispersion of the static magnetic susceptibility in wave vector space, respectively. This equation is regarded as the Landau expansion of the magnetic free energy F_m as follows:

$$F_m/N_0 = \frac{1}{2}r\left(\frac{p}{2}\right)^2 + \frac{\overline{F_1}}{4}\left(\frac{p}{2}\right)^4 \tag{3}$$

where r and $\overline{F_1}$ represent the second and the fourth order expansion coefficients of magnetic free energy, respectively. By comparing Eqs. (2) and (3), the parameter $\overline{F_1}$ is connected with the dynamical parameters T_0 and T_A by the following equation:

$$\overline{F_1} = \frac{4}{15} \frac{k_B T_A^2}{T_0} \quad (4)$$

The value of $\overline{F_1}$ and the saturation moment p_s are obtained experimentally from the Arrott plot analysis. From Eq. (2) by substituting $h=0$, p_s is connected with T_0 and T_A by the following equation:

$$\frac{p_s^2}{4} = \frac{15\,T_0}{T_A} c\eta^4 \quad (5)$$

T_0 and T_A can be also be expressed in the following way as function of T_C, p_s and ratio $\overline{F_1}/k_B$

$$T_0^{5/6} = 8(15)^{1/2} c p_s^{-2} \left(\frac{\overline{F_1}}{k_B}\right)^{-1/2} T_C^{4/3} \quad (6)$$

From the above expressions, we readily see that upon macroscopic measurements such as magnetization and the Arrott-plot analysis, giving access to the ratio $\overline{F_1}/k_B$, p_s and the knowledge of T_C, we can estimate quantitatively the values of the energy scale of the spin fluctuations spectrum. The parameters of Co_2TiGa thus obtained are summarized in Table 1 with the parameters of MnSi, $ZrZn_2$, Ni_3Al and Sc_3In. The parameters of MnSi, $ZrZn_2$, Ni_3Al and Sc_3In were obtained directly by dynamical measurements such as neutron scattering and/or NMR relaxation measurement. It should be noted that the value of η for Co_2TiGa is almost equal to those of MnSi and $ZrZn_2$. As shown in the Table 1, the very weak itinerant electron ferromagnets are defined as those having very small values of η. On the same context, magnets with localized magnetic moments have always large η values because T_0 is about the same magnitude with T_C. And, for the weak itinerant electron ferromagnets, the following qualitative magnetic behaviors have been predicted by the Takahashi theory depending on the value of η: (1) For small η, the linear relation in Arrott-plots holds well over a wide temperature range, whereas, for large η, this is no longer valid at finite temperature. (2) For small η, the squared spontaneous magnetization behaves as $T^{4/3}$ while, as η increases, it becomes to obey rather the T^2 -dependence. These theoretical predictions are in good agreement with the behavior for Co_2TiGa. Namely, it is likely that the electronic state of Co_2TiGa lies between the state of the very weak itinerant electron ferromagnetic limit and the intermediate case such as Fe and Ni, where in the very weak itinerant electron ferromagnetic limit, only the small q-components of spin fluctuations play the predominant roles and the electronic state of Fe and Ni is in the intermediate one between the very weak itinerant electron ferromagnetic limit and a local moment one. It should be noted that $\sigma_s^2(T)$ of $ZrZn_2$ decreases linearly as function of T^2 [14]. Lonzarich and Taillefer also gave the quadratic temperature dependence of $\sigma_s^2(T)$ over a wide range well below T_C for weak itinerant electron ferromagnetic metals in their model, taking account of both longitudinal and transverse spin fluctuations [15].

Table 1
Characteristic parameters for the spin fluctuations in Co_2TiGa[a]

	$\overline{F_1}/k_B$ (K)	T_0 (K)	T_A (K)	η
Co_2TiGa	2.1×10^4	834	8.00×10^3	0.53
MnSi	8.2×10^3	231	2.08×10^3	0.51
$ZrZn_2$	1.3×10^4	321	8.83×10^3	0.38
Ni_3Al	1.3×10^5	3590	3.09×10^4	0.23
Sc_3Jn	1.6×10^5	565	1.18×10^4	0.21

[a] The parameters of others typical weak itinerant electron ferromagnets are also summarized from Ref. [11] to make a comparison.

Fig. 5 shows initial permeability μ vs. temperature curves for Co_2TiGa at various pressures. Initial permeability decreases rapidly just below the Curie temperature and then takes a nearly constant value with further rise in temperature. The Curie temperature was defined as the point of intersection of linear extrapolations from the high and low temperature ranges. The Curie temperature was found to shift from 128 K at ambient pressure to 114 K under a pressure of 11 kbar. The pressure dependence of the Curie temperature is shown as the inset in Fig. 5. It is found that the Curie temperature of Co_2TiGa decreases linearly with increasing pressure. The value of $\partial T_C/\partial P$ is found to be -1.27 K/kbar. This value is in good agreement with that reported previously [9].

DiMasi et al. reported the relationship between the Curie temperature and the pressure derivative of the Curie

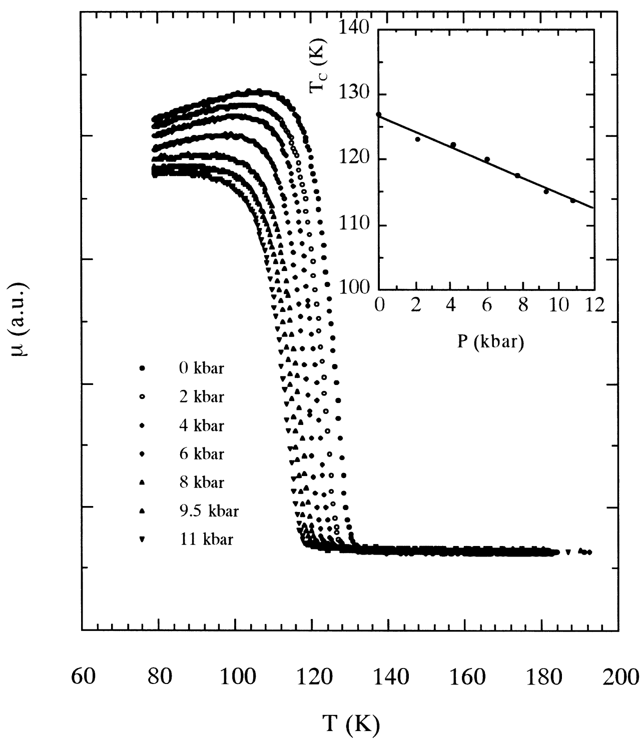

Fig. 5. Temperature dependence of the initial permeability μ for Co_2TiGa at various pressures. The inset shows the pressure change of the Curie temperature T_C for Co_2TiGa.

temperature for a number of itinerant electron magnets whose only potentially magnetic element is Co [10]. As seen in Fig. 6, the relationship between T_C and $\partial T_C/\partial P$ for materials having high Curie temperature is expressed by a universal curve. Using the collective electron model based on the molecular field theory, Wagner and Wohlfarth [16] derived the pressure dependence of the Curie temperature as follows,

$$\partial T_C/\partial P = (5/3)\,kT_C - A/T_C \quad (7)$$

where k is the compressibility, and A is a positive constant

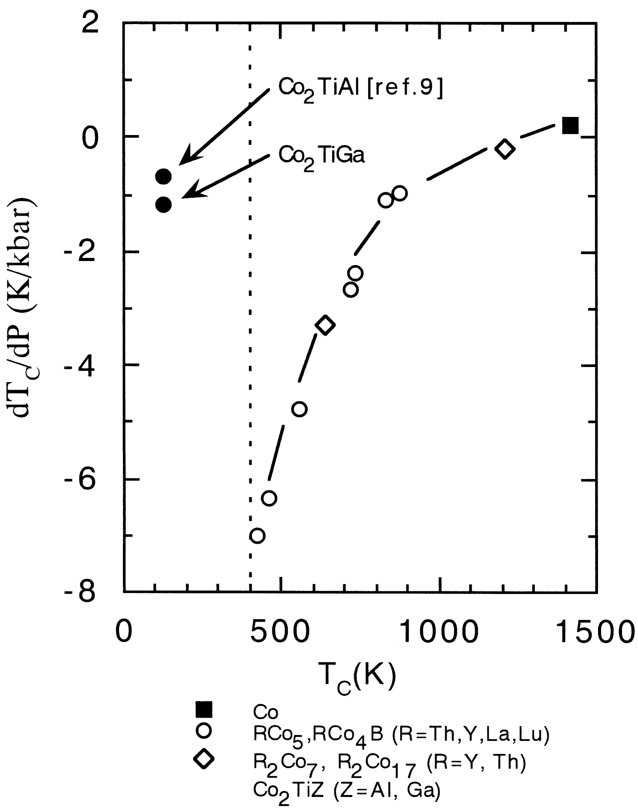

Fig. 6. Pressure derivative of the Curie temperature $\partial T_C/\partial P$ vs. the Curie temperature T_C for intermetallic materials whose only potentially magnetic species is Co. The line in the figure is a guide for the eye. This figure was first offered by DiMasi et al. [10] except for the present data of Co_2TiGa.

which its magnitude is concerned with the electronic structure of materials. It should be emphasized that the universal curve in Fig. 6 is expressed by the relation of $\partial T_C/\partial P \cong -A'/T_C$ (A' = constant). The second T_C^{-1} term in Eq. (7) comes from the contribution of d-band widening by pressure. As seen in Fig. 6, the tendency for cobalt Heusler alloys is less strongly pressure dependent than one would expect from extrapolation from the universal curve. This result may imply the presence of magnetic excitation such as the spin fluctuation in Co_2TiGa unaccounted for in the Wagner and Wohlfarth theory. We have noted above that the prediction of the spin fluctuation theory is in good agreement with the results for Co_2TiGa. Therefore, the magnetovolume effect for Co_2TiGa should be discussed in terms of the spin fluctuation theory. Now, the study of the pressure change of the magnetic moment is in progress for Co_2TiGa.

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