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Pressure-induced magnetic phase transition in CrTe at approximately 7 GPa

M. Ishizuka[a,*], H. Kato[a], T. Kunisue[a], S. Endo[a], T. Kanomata[b], H. Nishihara[c]

[a] *Research Center for Materials Science at Extreme Conditions, Osaka University, Toyonaka, Osaka 560-8531, Japan*
[b] *Faculty of Engineering, Tohoku Gakuin University, Tagajo 985-8537, Japan*
[c] *Faculty of Science and Technology, Ryukoku University, Seta 1, Otsu 520-2194, Japan*

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Abstract

High-pressure ac susceptibility and magnetization measurements have been made for the itinerant ferromagnet $Cr_{48}Te_{52}$ up to 8.5 and 17.2 GPa, respectively, to investigate the existence of a pressure-induced magnetic phase transition. The Curie temperature T_C (342 K at ambient pressure) decreased with increasing pressure at a rate of -53 K/GPa together with the height of the susceptibility peak associated with the paramagnetic–ferromagnetic transition. On applying pressures of 5–7 GPa, T_C decreased to about 70 K, and at pressures above 7 GPa any anomaly was not observed in the temperature dependence of the susceptibility. Magnetization measurements at low magnetic fields showed hysteresis of the M–T curves at low temperature which disappeared at about 7 GPa. These results suggest that the pressure-induced magnetic phase transition occurs at about 7 GPa in $Cr_{48}Te_{52}$. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Transition metal compounds; Magnetic measurements; Magneto-volume effect; High pressure; Phase transition

1. Introduction

Chromium telluride, $Cr_{1-\delta}Te$, is a ferromagnetic compound with metallic conductivity. Its Curie temperature T_C is 330–350 K, almost independent of the composition for $\delta \leq 0.25$, and T_C decreases largely for $\delta > 0.25$ [1,2]. There are six NiAs-related phases in the composition range of $\delta \leq 0.25$, which basically consist of an alternating stack of two kinds of metallic layers, those fully and partly filled with Cr ions [3]. The existence of stoichiometric CrTe is ruled out according to the results of the metallographical study of the Cr–Te system [3]. The crystal structure of the hexagonal NiAs-type is stable in the composition range between 52.4 ($\delta = 0.09$) and 53.5 ($\delta = 0.13$) at% Te. The amount of vacancies at Cr sites is an important quantity which characterizes the magnetic and the electronic properties together with the crystal structure [4,5].

As $Cr_{1-\delta}Te$ has large magneto-volume effects, many investigations have been carried out concerning magnetic properties as a function of atomic spacing [1,2,6–9]. In particular, their large negative pressure derivative of T_C, -70 K/GPa [2], has attracted researchers to a survey of a pressure-induced magnetic phase transition in $Cr_{1-\delta}Te$ due to a strong instability of the ferromagnetic state under pressure [7,8]. In 1973, Shanditsev et al. [7] measured the line width of the ESR absorption spectra for CrTe at various pressures up to 5 GPa to check the theoretical predictions made by Bean and Rodbell. At pressures above 3 ± 0.4 GPa, they could no longer detect the resonance line of the ferromagnetic phase in the temperature range between 100 and 300 K, which they attributed to a change in the nature of the magnetic state. Six years later, a neutron diffraction study on CrTe was carried out by the same group up to 3.5 GPa [8]. From a detailed comparison of the diffraction patterns obtained at various pressures and temperatures, they claimed the occurrence of a magnetic phase transition involving the disappearance of ferromagnetism at pressures above 3 GPa. There is, however, still room for doubt whether a magnetic phase transition occurs in the pressure range investigated in their experiments because their sensitivity seems insufficient, and the estimation of the ferromagnetic contribution from the diffraction pattern is very delicate due to a partial superposition of the spectrum of the material of the high-pressure cell and that of the CrTe compound. Recently, Takagaki et al. [10] calculated the electronic band structure of NiAs-type CrTe as a function of lattice parameters using a self-consistent

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*Corresponding author.
E-mail address: ishizuka@rcem.osaka-u.ac.jp (M. Ishizuka).

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LAPW method and showed the possibility of a pressure-induced ferromagnetic–antiferromagnetic transition.

In this work, we have made high sensitive magnetic measurements for $Cr_{1-\delta}Te$ with δ=0.077 ($Cr_{48}Te_{52}$) under high pressures up to 17.2 GPa to investigate the existence of the magnetic phase transition in this compound. The pressure dependence of T_C was obtained up to 8.5 GPa from the measurement of ac susceptibility χ_{ac} in a high-pressure cell loaded by a 250-ton press. For the investigation at higher pressures, a diamond-anvil cell (DAC) was employed and the temperature variation of the magnetization was measured at a magnetic field of 3 Oe using a superconducting quantum interference devices (SQUID) vibrating coil magnetometer (VCM).

2. Experimental

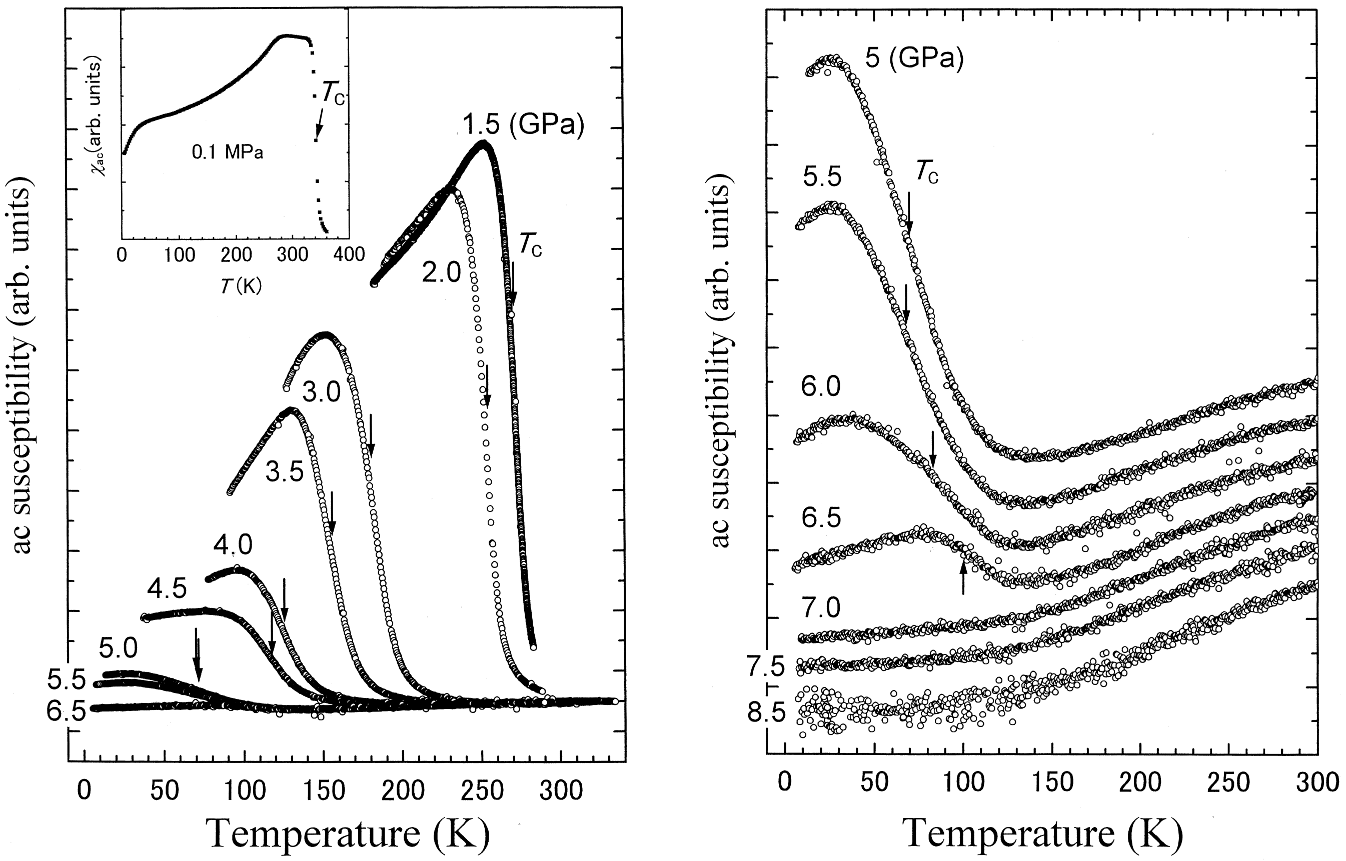
We have prepared polycrystalline samples of $Cr_{48}Te_{52}$ by a ceramic method using powders of Cr (99.9% purity) and Te (99.9999% purity). Powder X-ray diffraction measurements showed the compound obtained to be a single phase of NiAs-type crystal structure with no vacancy-order at the Cr site. For the measurement of χ_{ac}, a tightly compressed powder of the compound was placed in a Teflon capsule with Fluorinert introduced as the pressure medium. Primary and secondary coils were wound directly around the sample and the signal was detected by a lock-in-amplifier. The measurement frequency was 923 Hz. The capsule was set in the center of the pyrophyllite cube, which was pressurized uniformly by six anvils. The anvils and the compression rods were contained in a cryostat for variation of the temperature from around 4.2 K to room temperature. The load was controlled so that the pressure extended on the sample did not change during the measurement.

Magnetization measurements in a DAC were made using a technique of VCM combined with a SQUID [11]. Because the detection coil made of a superconducting wire vibrates just above the gasket of the DAC, the maximum temperature of the measurement is about 70 K, above which radiation from the DAC heats up the detection coil enough to destroy the superconductivity. A small amount of the compound and a chip of Pb used as a manometer were fixed in the hole of the CuBe gasket. A mixture of 4:1 methanol–ethanol was used as a pressure medium.

3. Results and discussion

Fig. 1a shows the temperature dependence of $\chi_{ac}(T)$ at pressures from 1.5 to 6.5 GPa. The inset shows $\chi_{ac}(T)$ at ambient pressure, measured at a frequency of 20 Hz using a commercial SQUID magnetometer. As shown in the figure, $\chi_{ac}(T)$ has a broad maximum just below T_C. Here, T_C was assigned as the inflection point of the $\chi_{ac}(T)$ curve, indicated by an arrow in the figure, taking into account the fact that the imaginary part of the susceptibility had a sharp peak at the corresponding temperature. In addition to a good agreement with the values reported in previous studies [2], the obtained T_C, 342 K, is in accordance with that determined from the measurement of the thermal expansion, 350 K, the coefficients of which change as a result of the spontaneous increase in the volume of the unit cell below T_C. When pressure is applied to the compound, T_C and the height of the peak associated with the paramagnetic–ferromagnetic transition decrease considerably, and at a pressure of 6.5 GPa, $\chi_{ac}(T)$ hardly changes with temperature. In view of the fact that the transition temperature obtained from this measurement decreases monotonically along the curve which almost fits to the pressure dependence of T_C reported previously, as shown below, the magnetic transition under high pressure may be considered to be the ferromagnetic one though the detailed nature of the low temperature phase is not clear. Magnified details of $\chi_{ac}(T)$ in the pressure range from 5 to 8.5 GPa are shown in Fig. 1b. At pressures above 5 GPa, T_C stays at around 70 K and seems to increase slightly with pressure. Considering that the temperature dependence of the background contribution to the susceptibility is almost the same for all the data, as shown in Fig. 1b, the peak disappears at around 7.0 GPa and any anomaly cannot be seen at higher pressures.

M–T curves at ambient pressure were measured at a magnetic field of 3 Oe in the temperature range from 4.2 to 360 K to obtain reference data for the high-pressure magnetization measurements at such a low magnetic field. The measurements were made using the same commercial magnetometer as that used for the measurement of $\chi_{ac}(T)$, and the results of zero-field cooling (ZFC) and field cooling (FC) in a magnetic field of 3 Oe are shown in Fig. 2. As shown in the figure, the paramagnetic–ferromagnetic transition is confirmed in both the curves, and the temperature dependence of $M_{ZFC}(T)$ is very similar to that of $\chi_{ac}(T)$ at ambient pressure. It appears that the compound exhibits spin–glass behavior below about 30 K at such a low magnetic field, which may be attributed to the antiferromagnetic freezing of the ferromagnetic clusters due to the random order of the vacancies at the Cr site [12]. Fig. 3a,b show typical examples of the results of the high-pressure magnetization measurements performed at temperatures ranging from 10 to 65 K. To achieve higher pressures, we used a smaller sample for the measurements shown in Fig. 3b because the CuBe gasket deforms so much with increasing pressure that the available size of the sample strongly depends on the value of the highest pressure of the measurement. The background contribution from the DAC was not subtracted from the results obtained for both samples. A large hysteresis is observed in the M–T curves at low pressures, suggesting the existence of the glassy phase observed at ambient pressure. It is seen from these figures that the temperature dependence of the



(a) (b)

Fig. 1. (a) Temperature dependence of ac susceptibility $\chi_{ac}(T)$ for $Cr_{48}Te_{52}$ at pressures from 1.5 to 6.5 GPa. Arrows show T_C, assigned as the inflection point of $\chi_{ac}(T)$. Inset shows $\chi_{ac}(T)$ at ambient pressure, measured at a frequency of 20 Hz using a commercial SQUID magnetometer. (b) Magnified details of $\chi_{ac}(T)$ at pressures ranging from 5 to 8.5 GPa. Baselines of the curves are shifted slightly to distinguish one from the other.

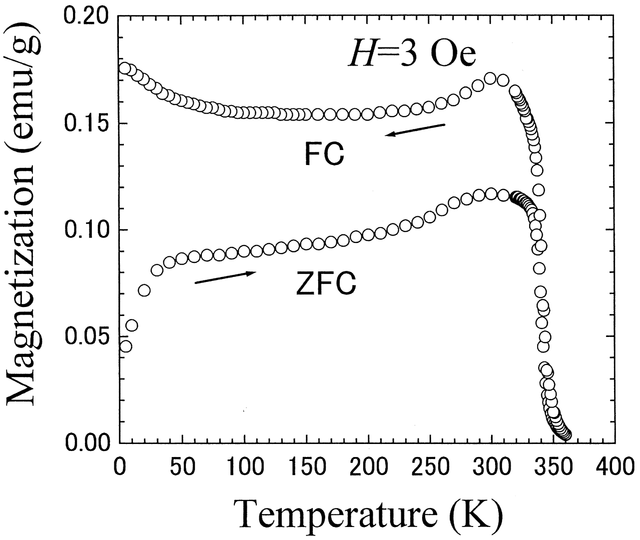
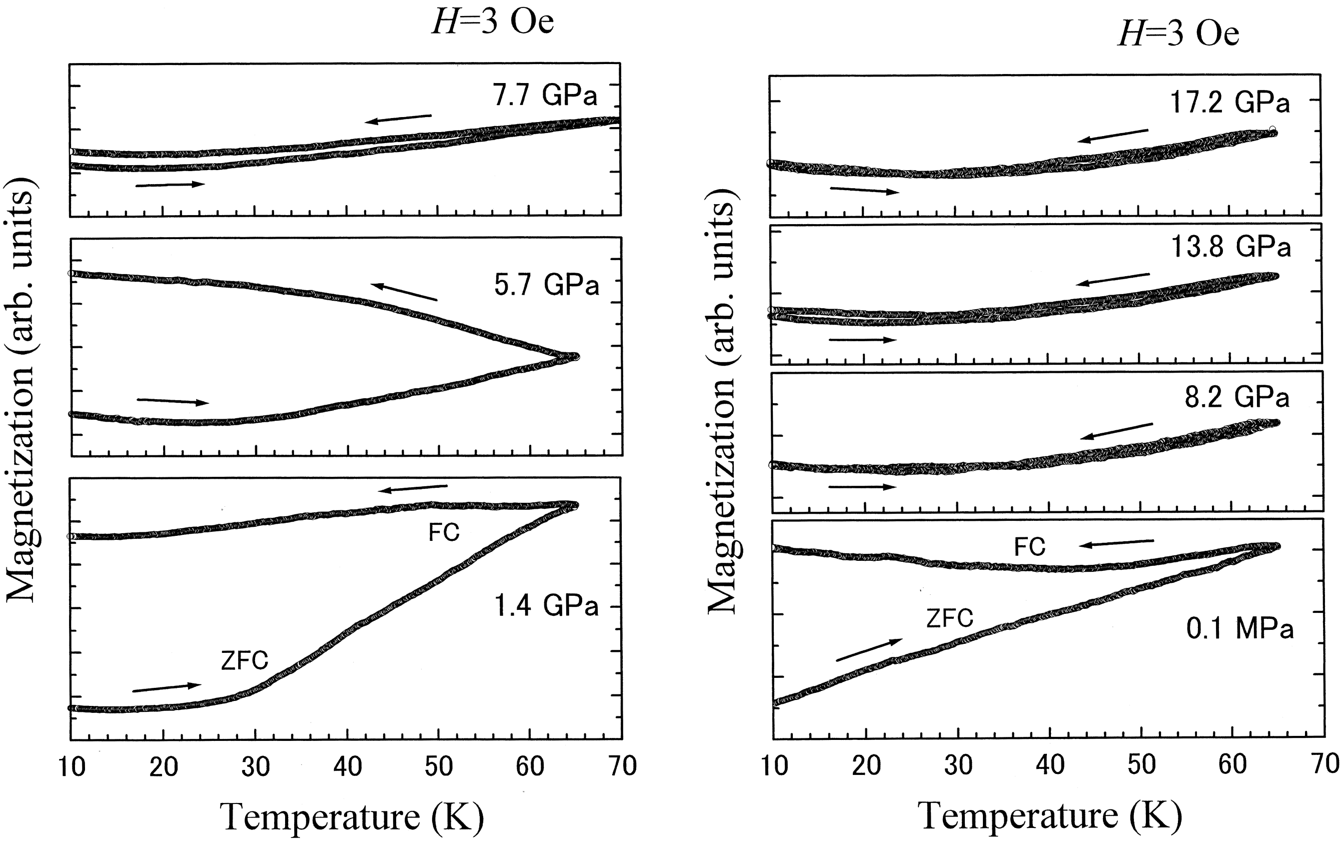


Fig. 2. Results of zero-field cooling (ZFC) and field cooling (FC) at ambient pressure and a magnetic field of 3 Oe.

magnetization together with the hysteresis of the M–T curves decreases with increasing pressure. At pressures above about 7 GPa, no appreciable change is observed in the curves. Fig. 4 shows the results of nine runs for different samples. For each run, measurement pressures are indicated by solid and open squares, corresponding to existence and absence of the hysteresis in the M–T curves, respectively. Half-solid squares indicate weak but significant hysteresis. As shown in the figure, the hysteresis of the M–T curves, evidence of the existence of the ferromagnetic interaction in this compound, is confirmed in the same pressure range where the magnetic transition was observed in the susceptibility measurement.

Fig. 5 shows the magnetic phase diagram of $Cr_{48}Te_{52}$ obtained from these measurements. Previous results [2,7] are included in the figure. As the freezing temperature of the ferromagnetic clusters was not measured as a function of pressure, the glassy phase is not included in the diagram. At pressures below 5 GPa, T_C decreases almost linearly with pressure at a rate of −53 K/GPa and reaches about 70 K. As shown in the figure, the pressure dependence of T_C is in good agreement with that of the previous studies, and besides, the measured points at higher



(a) (b)

Fig. 3. Typical examples of the results of $M_{ZFC}(T)$ and $M_{FC}(T)$ from 10 to 65 K at various pressures and a magnetic field of 3 Oe. Background contribution from the DAC was not subtracted from the results obtained for both samples. (b) Result for a smaller sample used to achieve higher pressures.

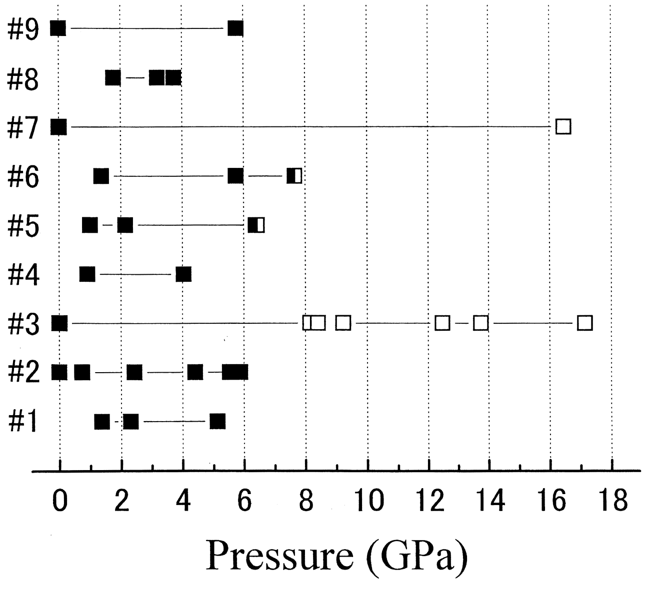


Fig. 4. Pressure range of the measurements of the magnetization for nine runs. Measurement pressures are indicated by solid and open squares, corresponding to existence and absence of the hysteresis in the M–T curves, respectively. Half-solid squares show weak but significant hysteresis.

pressures are on the line of their extension. This fact, together with the existence of the hysteresis of the M–T curves (hatched region in Fig. 5), suggests that the ferromagnetic state is stable in this pressure range and a pressure-induced magnetic phase transition occurs at about 7 GPa. As the high-pressure and low-temperature X-ray diffraction measurement shows that the NiAs structure is stable in this pressure and temperature range [15], the disappearance of the ferromagnetic behavior is not associated with a structural change in this compound. The previous studies claimed that the pressure-induced magnetic phase transition from ferromagnetic to antiferromagnetic or nonmagnetic states occurred at around 3 GPa, but there is no significant change in these results for $\chi_{ac}(T)$ and M–T curves at around 3–4 GPa. The discrepancy between the results of this and the previous studies may be attributed to the nature of the compounds used in the experiments: for the compound used in the previous studies, the neutron diffraction patterns at ambient pressure showed ordering of the Cr-vacancies and the associated antiferromagnetic phase appearing at low temperature [8]. This suggests that the compound mainly consists of Cr_3Te_4 ($\delta = 0.25$), which has the pseudo-NiAs structure with ordering of the Cr-vacancies and which exhibits a canted

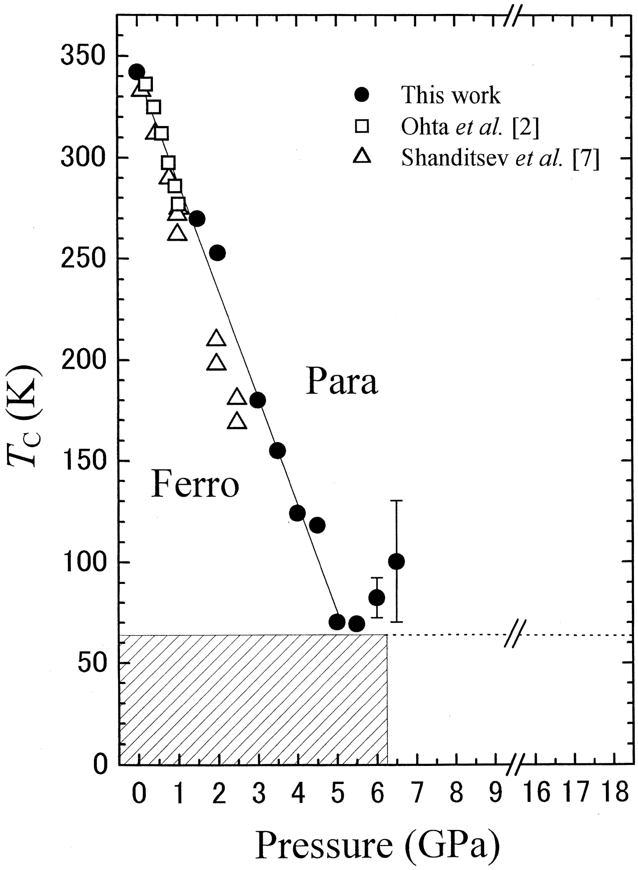


Fig. 5. Magnetic phase diagram of $Cr_{48}Te_{52}$. Open squares and triangles are from Refs. [2] and [7], respectively. Solid line is a visual guide. Horizontal dashed line is the upper limit of the magnetization measurement reached in this experiment, and the ferromagnetic behavior was confirmed in the hatched region.

ferromagnetic state below 80 K [13,14]. According to the theoretical calculation, the antiferromagnetic state is energetically stable compared with nonmagnetic and ferromagnetic states for the lattice parameter $a<3.83$ Å, and a ferromagnetic–antiferromagnetic transition is expected at about 20 GPa, which is more than 2 times the experimentally obtained value. The difference in the values of the critical pressure may arise mainly because the theoretical calculation was carried out with the ratio c/a fixed at the value obtained at ambient conditions. The magnetism in the high-pressure phase is, however, not clear in the present study because the measurement systems used in this work do not have sufficient sensitivity to investigate the magnetic state after the ferromagnetic behavior disappears. Further development of the technique for the measurement, especially for neutron diffraction experiments at very high pressures, will be needed for a detailed investigation of the magnetic phase transition in this compound.

4. Conclusions

From the measurement of ac susceptibility, T_C of $Cr_{48}Te_{52}$ was found to decrease with increasing pressure at a rate of -53 K/GPa and to reach about 70 K at 5 GPa. At pressures above 7 GPa, the height of the susceptibility peak decreased remarkably and the hysteresis of the M–T curves at low temperature disappeared, suggesting that a pressure-induced magnetic phase transition occurs around this pressure.

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