Magnetization Process near the Curie Temperature of an Itinerant Ferromagnet CoS$_2$

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Abstract. The field dependence of the magnetization of a typical itinerant ferromagnet CoS$_2$ has been reinvestigated close to the Curie temperature of 119.5 (0.5) K up to an applied field of 50 kOe. The critical index $\delta$ has been determined to be 5.2 (0.20). The result is in agreement with the theory by Takahashi for weak itinerant electron ferromagnets rather than the conventional molecular field theory. Reliable method for determining critical indices is also discussed.

1. Introduction

Self-consistent renormalized (SCR) spin fluctuation theory by Moriya et al. has successfully explained various magnetic properties at finite temperatures of weakly itinerant ferromagnets such as ZrZn$_2$, Sc$_3$In, MnSi, or Ni$_3$Al [1]. In spite of small saturation moments ($p_s$) of these materials, magnetic susceptibility shows Curie-Weiss behavior with effective moment $p_{\text{eff}}$ much larger than $p_s$ (i.e. $p_{\text{eff}} / p_s \approx 1$). Later, Takahashi has developed a theory from the different point of view that assumes the conservation of the total spin amplitude on each magnetic ion that consists of zero-point and thermal components [2]. One of the prominent features of the theory is the prediction of the critical magnetic isotherm at the Curie temperature $T_C$. The theory, well-adapted for the case with small magnetic moment, shows that the fourth order expansion coefficient of the magnetic free energy in the magnetization $M$ vanishes at the Curie temperature and $M^4$ becomes proportional to $H / M$ [2] instead of the linear relation between $H / M$ and $M^2$ in the so-called Arrott plot. Such a critical magnetization process were actually observed in MnSi [2,3] and Fe$_x$Co$_{1-x}$Si [4], while the linear relation in the Arrott plot was observed to be better in the case of a Heusler alloy Co$_2$TiGa [5].

The static critical behavior near $T_C$ of pure nickel has been investigated by many workers for more than four decades; however, the results had been discussed based on localized models, such as short-range
Heisenberg and Ising models [6-9]. The critical index $\delta$ defined by

$$H = D \cdot M(T, H)^\delta \quad (T = T_C)$$  \hspace{1cm} (1)$$

was reported to be 4.22 - 4.58 for nickel [6-9], 4.0 for gadolinium [10], 2.78 for YFeO$_3$, and 5.8 for CrO$_2$ [11]. The values of $\delta$ in early reports seem to be less reliable because they depend strongly on the value of $T_C$, which was determined by assuming the linear isotherm in the Arrott plot passing through the origin [7, 10]. The method is not self-consistent except for the case where $M^2$ versus $H/M$ plot becomes linear at $T_C$ i.e., $\delta = 3$. If the theory by Takahashi can be applied ($\delta = 5$), the $M^4$ versus $H/M$ plot becomes a straight line and passes through the origin at $T_C$. In later works, log ($M$) was plotted against log ($H$) and $T_C$ was determined as the temperature at which the plot became linear [9]. Theoretically, the molecular field theory and the SCR theory predict $\delta = 3$. They are also estimated in more sophisticated treatment for various models; 4.80 for the three-dimensional Heisenberg model [9], 4.82 for the three-dimensional Ising model [9], and 5.0 for itinerant electron ferromagnet [2].

Subsequent study has been reported on somewhat stronger ferromagnets, Ni$_2$MnGa and pure nickel, with higher Curie temperatures $T_C$ of about 363 and 623 K, respectively, and larger magnetic moments $p_s$ of 4.5 and 0.6 $\mu_B$/f.u., respectively [12]. The values of $\delta$ for the Ni$_2$MnGa and nickel samples were estimated to be 4.77 and 4.73, respectively, and they were interpreted to be close to the value of 5.0 due to the theory by Takahashi.

In view of the scattered values of $\delta$ in literatures, further studies were directed to other members of cobalt-based Heusler alloys; Co$_2$CrGa and Co$_2$VGa. The former alloy, Co$_2$CrGa is somewhat stronger ferromagnet with a saturation moment of 3.01 $\mu_B$/f.u. [13], where the values of $T_C$ and $\delta$ were determined to be 488.0 K and 4.93, respectively from the data of magnetization process near $T_C$, and the values of $\delta$ has been again interpreted to be close to the theoretical value, 5.0 predicted by Takahashi [14]. The latter, Co$_2$VGa is somewhat weaker ferromagnet with a saturation moment of 2.1 $\mu_B$/f.u. and $T_C$ of about 350K [15], where the critical index $\delta$ has been determined to be 4.15 which is an intermediate value between 5 and 3 [16]. It seems therefore that the larger $\delta$ around 5 tends to be observed for stronger ferromagnets, while the smaller $\delta$ deviated from 5 for weakly ferromagnetic Co-based Heusler alloys where Takahashi's theory is better to be applied.

The present experiment is therefore planned for a typical itinerant ferromagnet, CoS$_2$ with a moment of 0.84 $\mu_B$/Co and the Curie temperature of about 120 K, which are similar to those of Co$_2$TiGa. Static critical properties of CoS$_2$ was already investigated, and critical indices were obtained by fitting experimental isothermal magnetization curves to the expression proposed by Arrot and Noakes [7],

$$c_1 \left( \frac{H}{M} \right)^\gamma = \frac{T - T_C}{T_C} + c_2 M^\beta \delta = \frac{\gamma}{\beta} + 1$$  \hspace{1cm} (2)$$

where $\beta, \gamma$ are critical indices for the magnetization below $T_C$ and the paramagnetic susceptibility above $T_C$, respectively, and $c_1, c_2$ are numerical constants. Using this fitting, anomalously large value of 6.4 for $\delta$ was reported [17-19].
2. Experimental Results and Discussion

A SQUID magnetometer (Quantum Design MPMS-5S) was used for the measurements of the isothermal magnetization curves of CoS\(_2\) powder samples. The powdered sample was prepared in a conventional manner. Temperature derivative of the magnetization of CoS\(_2\) is shown as a function of temperature in Fig.1. From the figure, Curie temperature \(T_C\) is estimated to be about 119K, which is slightly lower than those reported in literatures [17-19].

![Fig.1. Temperature derivative of the magnetization of CoS\(_2\) as a function of temperature.](image1)

![Fig.2. \(M^2\) is plotted against \(H_{int}/M\) for CoS\(_2\).](image2)

![Fig.3. \(M^{5.4}\) versus \(H_{int}/M\) for CoS\(_2\) corresponding to \(\delta\) of 6.4.](image3)

![Fig.4. \(M^{4.2}\) versus \(H_{int}/M\) for CoS\(_2\) corresponding to \(\delta\) of 5.2.](image4)

Observed isothermal magnetization curves are shown in Fig.2, where \(M^2\) is plotted against \(H_{int}/M\). The externally applied field up to 50 kOe was transformed to the effective internal field \(H_{int}\) using the demagnetizing factor which was determined from the magnetization data in fields less than 100 Oe. If the molecular field theory with \(\delta\) of 3.0 is valid, the curve becomes a straight line at around 119K, but
apparently, this is not the case. In Fig.3, $M^{5/4}$ versus $H_{int}/M$ is plotted for CoS$_2$ corresponding to $\delta$ of 6.4 reported in the literatures [17-19]. It is clear that the curves around 119K are not straight lines but is slightly concave. On the other hand, the fairly good linearity of the curve for $\delta=5.2$ is observed as shown in Fig. 4. From the present analysis, the Curie temperature of 119.5 (0.5) K and critical index $\delta$ of 5.2 (0.20) are estimated. It also suggests that the analysis by fitting experimental data to the equation (2) with 5 adjustable parameters does not seem to be reliable. We therefore conclude that the observed isothermal magnetization curve of CoS$_2$ at the Curie temperature is well described by the smaller index $\delta=5.2$ than the previous ones, close to the value 5.0 by Takahashi for weak itinerant ferromagnets.

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