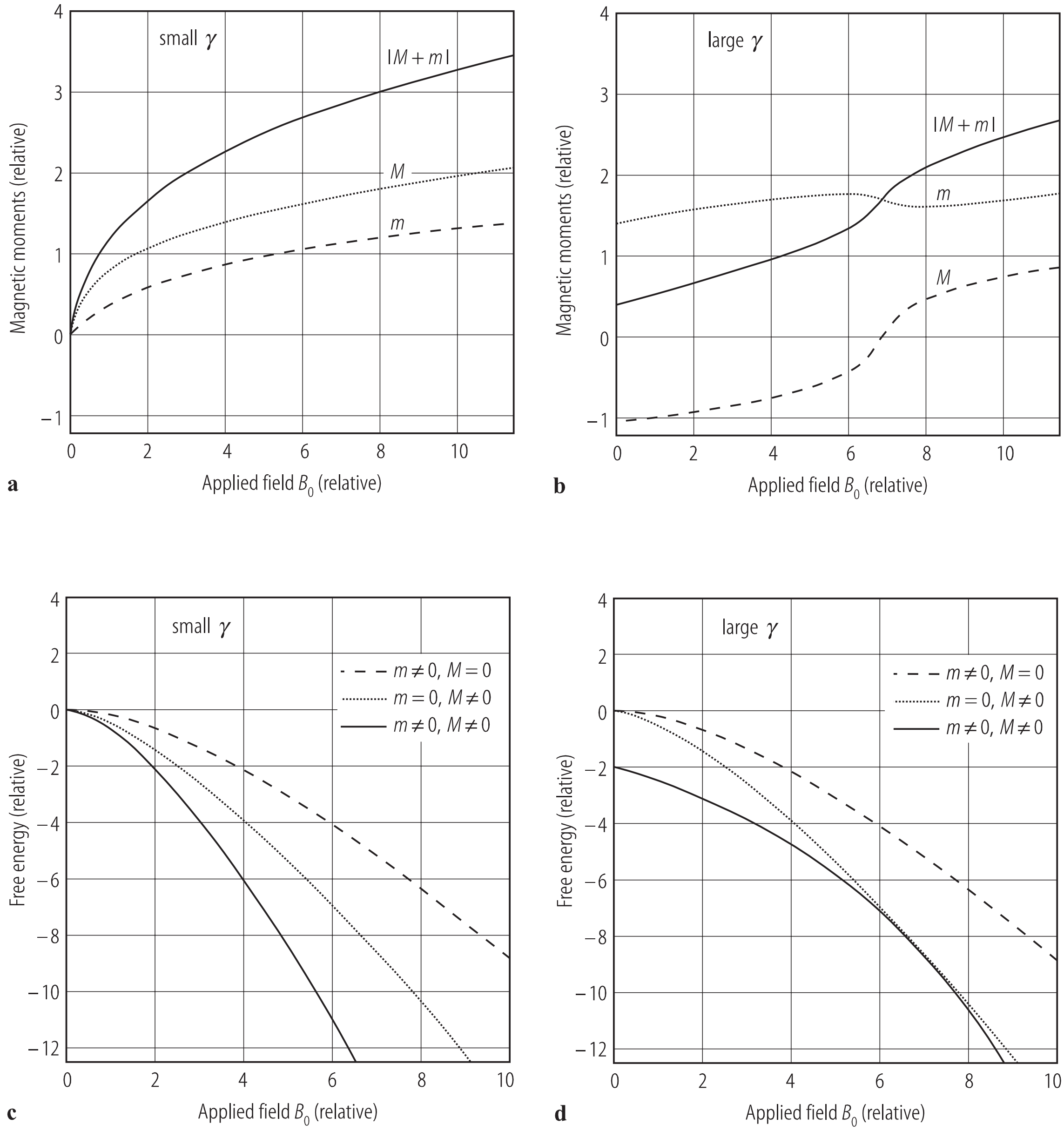

Fig. 66. Magnetic moments (M: dotted line, m: dashed line, $|M + m|$: full line) for small (**a**) and large (**b**) coupling constants γ. The corresponding free energies are shown in (**c**) and (**d**) for small and large γ values, respectively. The dashed (dotted) curves in (**c**) and (**d**) correspond to the free energy with $m \neq 0$, $M = 0$, ($M = 0$, $M \neq 0$) and the full line to $m \neq 0$, $M \neq 0$.

1.5.5.3.2.5 Ferromagnetic and / or antiferromagnetic order

Here the situation of more than one magnetic interaction present in the sample is considered. For a structure with only one magnetic sublattice and taking into account only one ferromagnetic moment $\boldsymbol{M}$ and an antiferromagnetic one $\boldsymbol{L}$, the free energy may be written in the form

$$F = \frac{1}{2}\tilde{A}\, M^2 + \frac{1}{4} C\, M^4 - \boldsymbol{B}_0 \cdot \boldsymbol{M} + \frac{1}{2}\, \tilde{a}\, L^2 + \frac{1}{4} c\, L^4 + \frac{1}{2}\, \gamma L^2 M^2 + \delta(\boldsymbol{L} \cdot \boldsymbol{M})^2 \tag{52}$$

The contributions proportional to γ and δ are the lowest order coupling terms between $\boldsymbol{M}$ and $\boldsymbol{L}$ which are always allowed by symmetry. The term proportional to δ will determine the orientation of the antiferromagnetic moment with respect to the ferromagnetic one. For $\delta > 0$ the free energy is minimised for $\boldsymbol{M} \perp \boldsymbol{L}$ and $\delta(\boldsymbol{L}\cdot\boldsymbol{M})^2 = 0$, while for $\delta < 0$ a collinear arrangement with $\boldsymbol{L} || \boldsymbol{M}$ will be preferred. With these remarks $\boldsymbol{M}$ will be oriented parallel to the external magnetic field direction and $\boldsymbol{L}$ will have a direction with respect to $\boldsymbol{M}$ as determined by the sign of the coefficient δ in eq. (52). The term proportional to δ in eq. (52) may therefore be incorporated into the term $\gamma \boldsymbol{L}^2\boldsymbol{M}^2$ with a renormalized coefficient γ. For the following it suffices to consider $\boldsymbol{M}$ and $\boldsymbol{L}$ as scalar quantities.

Minimising the free energy in eq. (52) with respect to both M and L one obtains

$$\frac{\partial F}{\partial M} = 0 \qquad \Rightarrow \; \tilde{A}\,M + CM^3 - B_0 + \gamma L^2 M = 0$$

$$\frac{\partial F}{\partial L} = 0 \qquad \Rightarrow \; \tilde{a}\,L + cL^3 + \gamma L M^2 = 0 \tag{53}$$

There are four different solutions for eq. (53), namely

$$(1) \quad M = 0 \quad L = 0 \qquad\qquad (2) \quad M = 0 \quad L \neq 0$$

$$(3) \quad M \neq 0 \quad L = 0 \qquad\qquad (4) \quad M \neq 0 \quad L \neq 0 \tag{54}$$

The coefficients A and a are assumed to have a temperature dependence given by

$$\tilde{A} = A'(T - T_C) \qquad\qquad a = a'(T - T_N) \tag{55}$$

(A', $a' > 0$) with C, c, A' and a' taken to be temperature independent. If the temperature is lowered and only one subsystem is considered, T_C (T_N) is the temperature for which the subsystem becomes unstable towards ferromagnetic (antiferromagnetic) order. As shown above the instability occurs at the temperature for which the A or a coefficients change sign as a function of temperature.

For $T > T_C$, T_N and with no external magnetic field applied the system is in its paramagnetic state. The inclusion of the coupling does not change this conclusion. Within this temperature range solution (1) of eq. (54) is realised.

As the temperature is lowered, magnetic order will set in for the magnetic order with the highest transition temperature. The sample will order ferromagnetically if $T_C > T_N$, and antiferromagnetically for $T_N > T_C$. Experimentally it is often found that the magnetic order (with the ferromagnetism and antiferromagnetism being examples thereof) with the highest transition temperature is also the magnetic order of the ground state. It is interesting to ask the question of what happens to the magnetic instability of the magnetic order with the lower transition temperature as the temperature is lowered even further.

In order to investigate this situation let $T_N > T_C$. With the assumption that the external magnetic field is small and the size of the ferromagnetic moment M is also small one may neglect the $\frac{1}{4}CM^4$ term in the free energy in eq. (52) and obtain the expression for M

$$M = \frac{1}{\tilde{A} + \gamma L^2}\, B_0 \tag{56}$$

The ferromagnetic instability will occur whenever the ferromagnetic susceptibility diverges. As seen from eq. (56) the $\tilde{A}$ coefficient is renormalized due to the antiferromagnetic order. For $\gamma > 0$ and L^2 being always positive the effective coefficient is $(\tilde{A} + \gamma\, L^2)$, which does not change sign at T_C but at some other temperature. If the coupling constant γ is large and positive, the ferromagnetic instability of the system is eliminated, and the antiferromagnetic order is stabilised down to the lowest temperatures. The renormalization of the ferromagnetic interactions is also seen in the magnetic susceptibility as measured in magnetisation experiments. There it is observed that the magnetic susceptibility shows a maximum at T_N and decreases for $T < T_N$. The susceptibility is given by the inverse of the $\widetilde{A}$ coefficient above T_N and by $(\widetilde{A} + \gamma L^2)^{-1}$ for $T < T_N$. The decrease below T_N is seen to arise here due to the renormalization of the $\widetilde{A}$ coefficient with the renormalization being due to the antiferromagnetic ordering. The renormalization is proportional to the antiferromagnetic order parameter squared. This observation is a well known textbook result (see for example Kittel [76C1]) which has been re-derived here using the formulation of Landau theory.

A similar renormalization of the antiferromagnetic coefficient $\widetilde{a}$ will occur due to a non-zero ferromagnetic moment M. This renormalisation can be observed experimentally by studying the antiferromagnetic phase transition in an external magnetic field. For $T < T_N$, $B_0 \neq 0$ and using the linearisation of M (i.e. neglecting terms proportional to C) so that $M = \chi B_0$ eq. (53) for L now reads

$$(\widetilde{a} + \gamma M^2)L + cL^3 = 0 \tag{57}$$

or

$$\left(a'\left(T - T_N\right) + \gamma \chi^2 B_0^2\right)L + c\,L^3 = 0 \tag{58}$$

The change of sign of the renormalized a coefficient is determined by $\overline{\alpha} + \left(a'\left(T - T_N\right) + \gamma \chi^2 B_0^2\right)$. For $\gamma > 0$ the antiferromagnetic transition temperature is shifted to lower temperatures with increasing external magnetic field. Thus the change of T_N as a function of applied ferromagnetic field (which does not couple directly to the antiferromagnetic order parameter L) is evidence for the renormalization of the antiferromagnetic parameter. This renormalization may be investigated by bulk measurements such a specific heat.

The properties of Arrott plots for a system with an antiferromagnetic phase transition is also readily deduced. For temperatures above the transition temperature the antiferromagnetic order parameter is zero. With the application of an external magnetic field a ferromagnetic moment is induced, and with $L = 0$ the plot of the magnetisation as given in eq. (44) will result in straight lines. As the temperature is lowered and antiferromagnetic order sets in, the ferromagnetic A coefficient is re-normalised due to the antiferromagnetic order parameter L as indicated in eq. (57). Thus for γ large and positive the temperature dependence of the intersection with the x axis is changed in the Arrott plots. While for $T > T_N$ the lines are displaced to the left as temperature decreases, this displacement may be inverted to a displacement to the right for γ large and $T < T_N$. This is illustrated in Fig. 68.

It is of interest to consider two different situations, depending on the strength of the coupling constant γ. For strong coupling the antiferromagnetic order re-normalises the ferromagnetic susceptibility in such a way that the increase of the ferromagnetic moment with field is reduced compared to the case without antiferromagnetic order. As a result of this renormalization the ferromagnetic as well as the antiferromagnetic moment do not change significantly with field for low field strengths (see Fig. 69a). However, the antiferromagnetic state becomes unstable for large fields, and the system switches in a first order transition from a predominately antiferromagnetic state to a purely ferromagnetic one. This is illustrated in Fig. 69b for the free energy at temperature T_0. The Arrott plots for such a system are shown in Fig. 70, where the transition is apparent from the jump in the magnetisation.

For weak coupling the ferromagnetic moment increases continuously as a function of applied field. The renormalization of the antiferromagnetic component results in a continuous reduction of this order parameter with increasing applied magnetic field (see Fig. 71a). The antiferromagnetic order parameter goes to zero, and at a critical applied field the free energies of the purely ferromagnetic and the mixed ferro-antiferromagnetic state become equal (Fig. 71b). The Arrott plots are shown in Fig. 72.

The magnetic behaviour as given above may be applied to systems with the orientation of the magnetic moments given by either $\boldsymbol{M} \parallel \boldsymbol{L}$ or $\boldsymbol{M} \perp \boldsymbol{L}$. Only a qualitative argument is put forward here to support this point of view.

Consider the ground state of the system at $T = 0$. The magnetic moments are all aligned, and for fixed moment systems, the size of the ordered component is maximal. For $\boldsymbol{M} \parallel \boldsymbol{L}$ an applied field will try to increase the size of the ferromagnetic moment. This, however, will involve quite large amounts of energy. As a consequence the increase of the ferromagnetic component with applied magnetic field is reduced as compared to the increase for the paramagnetic state. This is modelled in the free energy by a large coupling constant γ, and thus the situation as depicted in Fig. 69 and Fig. 70 is applicable.

For an orientation of magnetic moments as given by $\boldsymbol{M} \perp \boldsymbol{L}$, the applied magnetic field will essentially keep the magnitude of the magnetic moments fixed and begin to turn the antiferromagnetically aligned moments into the external field direction. Here the coupling of the ferromagnetic and antiferromagnetic component is relatively weak, and the behaviour is characteristic of the one shown in Fig. 71 and Fig. 72.

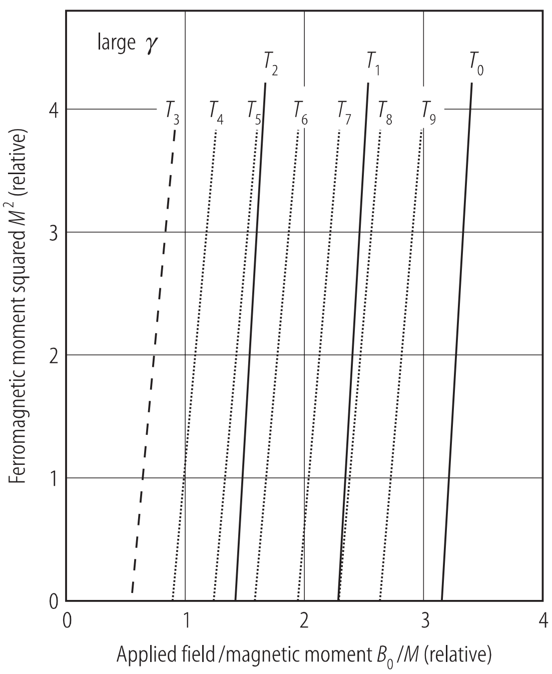

Fig. 68. Arrott plots for low external magnetic fields for a substance with antiferromagnetic order at T_3. As the temperature increases from T_0 to T_3 in the antiferromagnetic phase the Arrott plots are displaced to the left. In the paramagnetic phase the displacement is to the right for increasing temperatures. Due to the renormalization of the ferromagnetic properties by the antiferromagnetic order the slope of the Arrott plots changes.

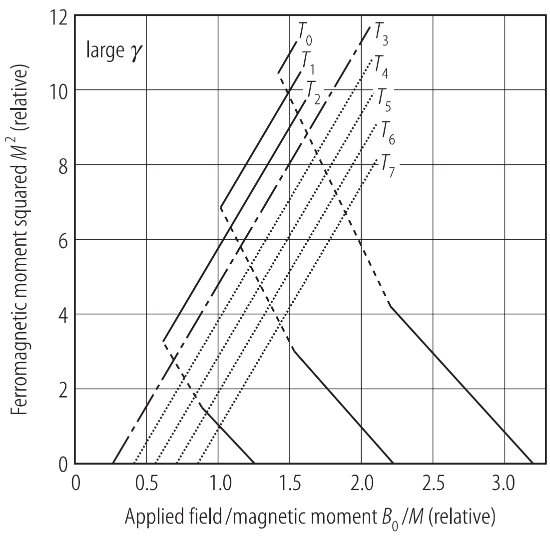

Fig. 70. Arrott plots for the case of strong coupling and the case illustrated in Fig. 69. The discontinuous increase of the ferromagnetic moment in the antiferromagnetically ordered state is indicated by the dashed line. The antiferromagnetic transition temperature is T_3, and the paramagnetic state is the thermodynamically stable one for temperatures T_4 to T_7.

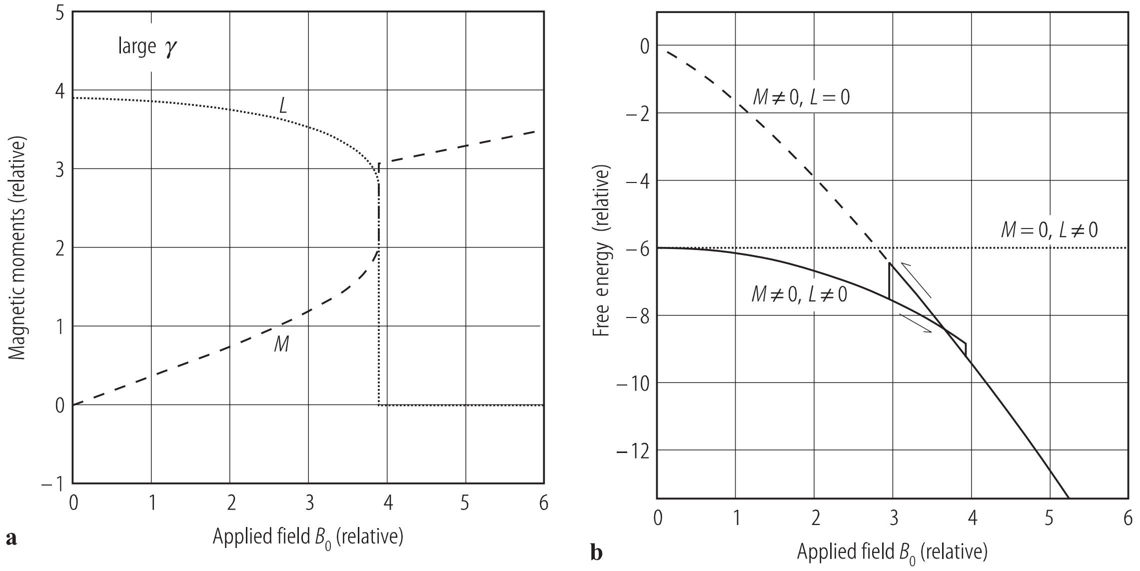

Fig. 69. In (**a**) the magnetic moments (antiferromagnetic moment L: dotted line, ferromagnetic moment M: dashed line) are shown as a function of applied magnetic field and for strong coupling. The corresponding free energy is shown in (**b**). A first order transition to a purely ferromagnetic state occurs at high magnetic fields. In (**b**) the hysteresis is shown for increasing and decreasing fields. The free energy for the antiferromagnetic state (dotted line, ferromagnetic moment M = 0) is field independent. The purely ferromagnetic free energy is indicated by the dashed line (antiferromagnetic moment L = 0). The full line corresponds to the (local) minimum of the free energy with both ferromagnetic and antiferromagnetic moments non-zero.

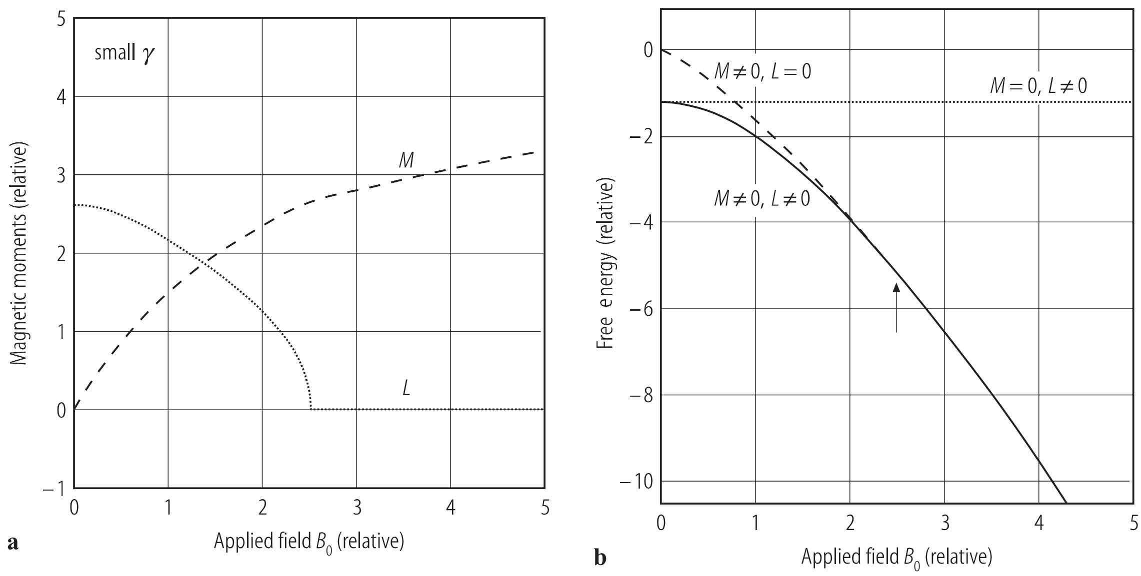

Fig. 71. Magnetic moments (**a**) and free energy (**b**) for the case of weak coupling. The significance of the symbols is the same as in Fig. 69. The arrow in (**b**) indicates the point at which the free energies of the full and dashed lines become equal.

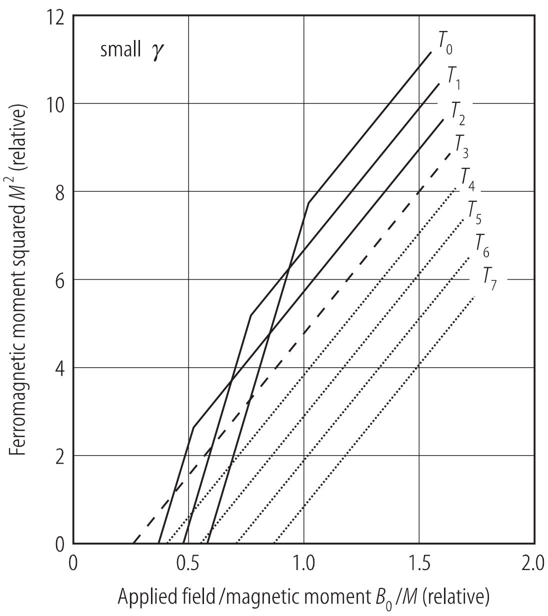

Fig. 72. Arrott plots for the case of weak coupling. The symbols and other parameters are identical to those of Fig. 70.

1.5.5.3.2.6 Discussion

The above analysis of some magnetic model systems has illustrated the potential of Landau theory and its application to the description of magnetic properties. With the help of Arrott plots the connection is made to the free energy. In favourable circumstances the parameters and their temperature dependence may be obtained from the experimental data by the use of Arrott plots. This allows a description of magnetic properties taking into account the knowledge of the system, and in particular its symmetry. If the symmetry of the magnetic structure is also known the modelling may be used to obtain a description of the effect of temperature and field on the stability of the arrangement of magnetic moments. For clarity, effects arising from fluctuations of the order parameter have been neglected. However, if important they can be incorporated into the present formulation in a straightforward manner. Other possible complications such as the presence of crystal fields have been dealt with elsewhere [95N3].

1.5.5.3.3 Experimental results

The majority of the magnetic properties have already been reviewed in [88W1]. Those which have appeared since are primarily more detailed examinations of the effect of atomic order or new compounds containing rare-earth elements.

1.5.5.3.3.1 Ferromagnets

X_2YZ X = 3d, Y = 3d

X = 8A: Ni; 1B: Cu
Y = 7A: Mn
Z = 3B: Al, Ga, In; 4B: Sn

Alloys in the series Cu_2MnZ and Ni_2MnZ are, with the exception of Ni_2MnAl, ferromagnetic [88W1]. The compounds form the Heusler $L2_1$ structure and a moment of approximately $4\mu_B$ is