# Magnetic properties of $Ni_3Al$ and $Ni_3Ga$: emergent states and the possible importance of a tri-critical point



2009 J. Phys.: Condens. Matter 21 095601

(http://iopscience.iop.org/0953-8984/21/9/095601)

J. Phys.: Condens. Matter **21** (2009) 095601 (5pp) doi:10.1088/0953-8984/21/9/095601

# Magnetic properties of $Ni_3Al$ and $Ni_3Ga$: emergent states and the possible importance of a tri-critical point

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Received 16 October 2008, in final form 13 January 2009
Published 4 February 2009
Online at stacks.iop.org/JPhysCM/21/095601

**Abstract**
We present a study of the magnetic properties of the itinerant-electron systems $Ni_3Al$ and $Ni_3Ga$ at ambient pressure. In both compounds the magnetization and susceptibility show a non-Fermi liquid form. We test these properties using a mean-field model of enhanced spin fluctuations on the border of ferromagnetism in three dimensions with no adjustable parameters. While $Ni_3Al$ is found to be explained well by the standard form of such a model, the data on $Ni_3Ga$ require us to extend the model to take into account the fact that this system lies close to a tri-critical point. We suggest that such a quantum tri-critical point may be a key feature in the understanding of quantum critical systems more generally.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

The standard model of metals, Landau Fermi liquid theory, has been used as the basis of understanding metallic properties at low temperatures for over 50 years. However, the properties of metals near to magnetic phase transitions at low temperatures are often found to deviate from those predicted by this standard model. In order to understand this deviation from Fermi liquid theory we need to consider relatively simple materials which show such behavior and seek a description of them using a simple model.

Some of the simplest systems which show a ferromagnetic to paramagnetic phase transition at low temperature are the d-electron systems, including $Ni_3Al$, $Ni_3Ga$, $ZrZn_2$, MnSi and $CoS_2$. The advantage of such systems is that they are itinerant in nature and have a small spin–orbit interaction. The theoretical analysis of such systems has a long history and was originally based on the so called Stoner–Edwards–Wohlfarth theory [1, 2] of itinerant ferromagnetism and later on a mean-field treatment of the effects of strongly enhanced spin fluctuations (para-magnons) [3–6]. This latter model is often called the self-consistent renormalization or SCR model. For a metal on the border of ferromagnetism in three spatial dimensions (3D) this model predicts a $T^{4/3}$ temperature dependence of the inverse susceptibility, compared to the conventional $T^2$ form expected from Landau Fermi liquid theory.

In this paper we examine the low-temperature magnetic properties of the ferromagnet $Ni_3Al$ and its close paramagnetic relative, $Ni_3Ga$. $Ni_3Al$ can be prepared in a pure stoichiometric form and crystallizes in a simple cubic ($Cu_3Au$) structure. At ambient pressure it orders ferromagnetically below 41 K with a small average moment of $0.075\mu_B$/Ni in the low-temperature, low magnetic field limit [7, 8]. The ferromagnetism is suppressed by the application of hydrostatic pressure and $Ni_3Al$ is found to become paramagnetic above 82 kbar [9]. By contrast $Ni_3Ga$, which has the same crystal structure and a similar electronic structure [10], lies on the paramagnetic side of the quantum critical point. Therefore, these two materials form a good basis on which to test the SCR model in both the ferromagnetic and paramagnetic state.

$Ni_3Al$ and $Ni_3Ga$ have been the subject of many investigations which show the importance of spin fluctuations in these systems. In particular, de Haas van Alphen studies [11–13] show the itinerant nature of the magnetism and neutron scattering measurements [14–16] show a para-magnon spectrum which is consistent with the SCR model and allow an evaluation of the model parameters.

In terms of the low-temperature magnetization and susceptibility, which concern us in this letter, past measurements of $Ni_3Al$ [7, 17] seem broadly consistent with the SCR model [18]. However, the limited measurements of the low-temperature inverse susceptibility of $Ni_3Ga$ [7, 19] appear to show a temperature dependence similar to $T^2$. This seems to be in contradiction to the SCR model which predicts a $T^{4/3}$ dependence. Therefore, in this paper we re-examine the low-temperature magnetization and susceptibility of high-quality (RRR $\geqslant$ 40) stoichiometric single crystals of $Ni_3Al$ and $Ni_3Ga$. Our main aim is to explore the possibility of understanding something other than a $T^{4/3}$ temperature dependence of the inverse susceptibility for a paramagnetic material near the border of ferromagnetism. To do this we compare these measurements directly with calculations based on the simple SCR model with no adjustable parameters. While we recognize the more recent theoretical works that show that there should be non-analytic corrections to certain aspects of the SCR model [20–22], we believe that we can still gain some important insight into the problem by a comparison with the basic SCR model. In particular, the major effects of these corrections are to alter the form of the zero-temperature equation of state and the momentum dependence of the susceptibility. In what follows both of these quantities are taken from experiment and we believe there is still value in using an SCR approach to calculate the effect of thermal fluctuations on the finite-temperature properties.

## 2. The self-consistent renormalization model

We now give the barest outline of the key ideas within the self-consistent renormalization (SCR) model in order to define the model and the model parameters so that we may compare its predictions directly with experiment. We focus on a paramagnetic system close to the border of ferromagnetism and follow the convention of [18].

It is assumed that in the low-$T$ limit the magnetization $\mathbf{M}(\mathbf{r})$ in a weak applied magnetic field $\mathbf{H}(\mathbf{r})$ is given by a Ginzberg–Landau equation of the form

$$\mathbf{H} = a\mathbf{M} + b\mathbf{M}^3 - c\nabla^2\mathbf{M}, \quad (1)$$

where $\mathbf{M}^3 = (\mathbf{M}\cdot\mathbf{M})\mathbf{M}$ and $a$, $b$ and $c$ are positive constants for a fixed pressure. We also assume that the relaxation of a fluctuation in the magnetization is controlled by Landau damping, i.e. that a Fourier component $\mathbf{M}_\mathbf{q}(t)$ of $\mathbf{M}(\mathbf{r}, t)$ decays exponentially to the value given by equation (1) via a relaxation function that is given by $\gamma q$ where $\gamma$ is a constant and $q = |\mathbf{q}|$. Thus, within this model the state of our system at zero temperature is defined by four parameters: $a$, $b$, $c$ and $\gamma$. The parameters $a$ and $b$ can be determined from the zero-temperature limit of the magnetization measurements as a function of magnetic field and $c$ and $\gamma$ can be extracted from inelastic neutron scattering measurements.

The temperature dependence of the magnetic equation of state is assumed to arise primarily from the effect of strongly enhanced long-wavelength spin fluctuations. The effect of these fluctuations, to lowest order in the paramagnetic state with no applied field, is to renormalize the linear coefficient in equation (1) (which represents the inverse uniform susceptibility), i.e.

$$a \rightarrow A(T) = a + \tfrac{5}{3} b\, \overline{\mathbf{m}^2}, \quad (2)$$

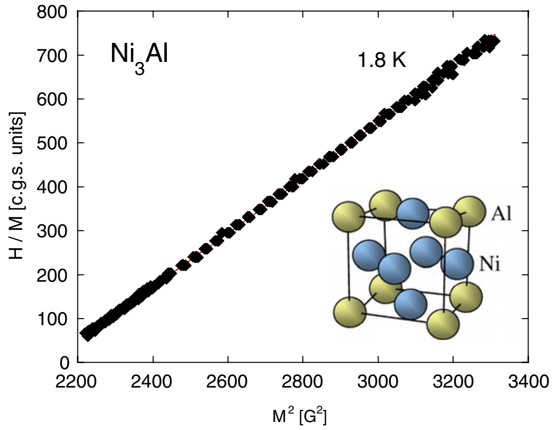


**Figure 1.** The main panel shows $H/M$ (the dimensionless inverse volume susceptibility in c.g.s. units of $Oe/(emu\ cm^{-3})$) versus $M^2$ for $Ni_3Al$ at 1.8 K for magnetic fields up to 70 kOe (7 T). The linearity of this plot demonstrates the applicability of the form of the equation of state given in equation (1) and allows determination of the parameters $a$ and $b$. The inset shows the simple cubic crystal structure of $Ni_3Al$ and $Ni_3Ga$.

where

$$\overline{\mathbf{m}^2} = \sum_q \overline{\mathbf{m}_q^2}. \quad (3)$$

$\overline{\mathbf{m}_q^2}$ is the thermal variance of a Fourier component of the fluctuating component of the magnetization defined by the fluctuation-dissipation theorem:

$$\overline{\mathbf{m}_q^2} = \frac{2\hbar}{\pi}\int_0^\infty \mathrm{d}\omega\, n_\omega\, \mathrm{Im}\, \chi_{q\omega}. \quad (4)$$

Here, $n_\omega$ is the Bose function and $\chi_{q\omega}$ is the wavevector and frequency-dependent susceptibility. Note that we have not included the zero-point contribution [18]. In the SCR model we have over-damped fluctuations defined by

$$\chi_{q\omega}^{-1}(T) = A(T) + cq^2 - \frac{\mathrm{i}\omega}{\gamma q}, \quad (5)$$

where the parameters $c$ and $\gamma$ can be estimated from inelastic neutron scattering measurements. Equations (2)–(5) can be solved self-consistently to give the temperature dependence of the inverse susceptibility, $\chi^{-1} = A$. We emphasize that the temperature dependence of the susceptibility is obtained using only the four, non-adjustable, zero-temperature parameters. In the ferromagnetic case the situation is slightly more complicated as we have to consider the longitudinal and transverse fluctuations separately; this situation is described elsewhere [18].

## 3. $Ni_3Al$

We first consider magnetization measurements performed on a high-purity single crystal of $Ni_3Al$ using an MPMS SQUID magnetometer. The sample had a resistivity ratio of 50; details of the sample preparation can be found elsewhere [11]. Figure 1 shows that the dependence of the magnetization,

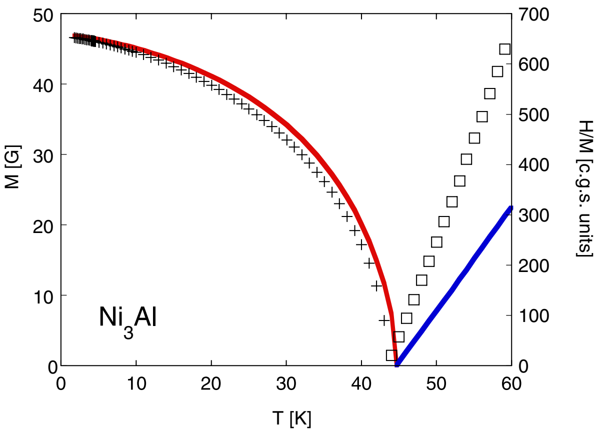


**Figure 2.** The zero-field magnetization, $M$, (crosses) and inverse volume susceptibility, $\chi^{-1}$, (squares) of $Ni_3Al$ versus temperature. These plots were derived by extrapolation of the linear fits to the Arrott plots at each temperature. The red and blue solid lines show the same quantities calculated within the SCR model using equations (2)–(5). The parameters $a = -1250$ and $b = 0.57G^{-2}$ were determined from the 1.8 K Arrott plot in figure 1. The parameters $c \approx 1.5 \times 10^5$ Å$^2$ and $\hbar\gamma \approx 3.3$ $\mu$eV Å were estimated from inelastic neutron scattering measurements [14].

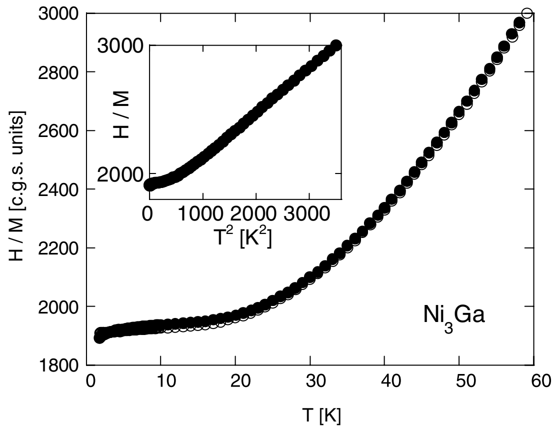


**Figure 3.** The dimensionless inverse volume susceptibility ($H/M$ in c.g.s. units of Oe/(emu cm$^{-3}$)) against temperature, $T$, at a field of 10 kOe (closed circles) and 1 kOe (open circles) for $Ni_3Ga$. The inset shows the same data plotted against $T^2$.

$M$, of $Ni_3Al$ on the applied magnetic field, $H$, in the low-temperature limit is well described by the equation of state given in equation (1). Here and below $H$ and $M$ refer to the magnitude of **H** and **M** as we assume an isotropic system. Using linear fits to the plots of $H/M$ against $M^2$ for each temperature we can obtain the temperature dependence of the zero-field magnetization, and the inverse susceptibility above $T_C$ as shown in figure 2. This shows the transition temperature to be just under 43 K. Also, from the linear inverse susceptibility at high temperatures we may derive the effective Curie–Weiss type moment to be around 17 times the low-temperature moment.

We can now compare this experimental data with the SCR model outlined above. Figure 2 shows the predictions of the model, with parameters relevant to $Ni_3Al$, plotted on top of the experimental data. For a model with no adjustable parameters the agreement is excellent. In particular, $T_C$ is predicted within 5% and the form of the magnetization and susceptibility are in good agreement. The main quantitative discrepancy is the slope of the inverse susceptibility above $T_C$. These broad conclusions were reached in an earlier comparison [18].

## 4. $Ni_3Ga$

We now move on to consider $Ni_3Ga$, the paramagnetic cousin of $Ni_3Al$. Magnetization measurements were also carried out on a high-purity single crystal of $Ni_3Ga$. The sample had a resistivity ratio of 40; details of the sample preparation can be found elsewhere [13]. The dependences of the inverse susceptibility with temperature at several fields up to 7 T were measured. In figure 3 we show the data for fields of 1 kOe (0.1 T) and 10 kOe (1 T). The fact that these two lie on top of one another means that below 1 T we are essentially in the low-field limit; this is assumed in the following analysis. According

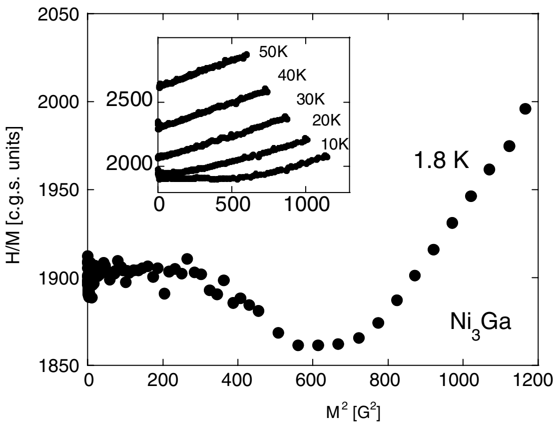


**Figure 4.** The main panel shows $H/M$ (the inverse volume susceptibility in c.g.s. units of Oe/(emu cm$^{-3}$)) versus $M^2$ at 1.8 K for magnetic fields up to 70 kOe (7 T). In contrast to $Ni_3Al$ the severe non-linearity of this plot shows that the equation of state given in equation (1) is not sufficient for $Ni_3Ga$. The inset shows Arrott plots at several higher temperatures, demonstrating that these plots become more linear at higher temperatures.

to the SCR model, the effect of thermal fluctuations on the zero-temperature equation of state, $H/M = a+bM^2$, is to give a dependence of the inverse susceptibility in the paramagnetic state given, for zero applied field, by equation (2). For a nearly ferromagnetic system such as $Ni_3Ga$, $\overline{\mathbf{m}^2}$ is predicted to vary with temperature as $T^{4/3}$, thus $\chi^{-1}$ should vary as $T^{4/3}$. However, as shown by the inset to figure 3, in this case the temperature dependence is much closer to a $T^2$ form. This result, which was seen in previous measurements, seems to be in sharp contradiction to the SCR model and is puzzling. A careful study of the low-temperature equation of state points toward a solution to this mystery. Figure 4 shows that the simple form of the equation of state given in equation (1), namely that $H/M = a + bM^2$, is not valid in this case. Therefore, we must include the next term in the expansion of the equation of state, such that:

$$\mathbf{H} = a\mathbf{M} + b\mathbf{M}^3 - c\nabla^2\mathbf{M} + g\mathbf{M}^5. \qquad (6)$$

If we now add in the effects of fluctuations in the same way as for the standard SCR model then the coefficients, $a$ and $b$, are renormalized in the following way:

$$a \rightarrow A(T) = a + \tfrac{5}{3}b\,\overline{\mathbf{m}^2} + \tfrac{35}{9}g\,\overline{\mathbf{m}^2}^2. \qquad (7)$$

$$b \rightarrow B(T) = b + \tfrac{14}{3}g\,\overline{\mathbf{m}^2}. \qquad (8)$$

So, if $b$ is small, $\chi^{-1}$, which is equal to $A$ in the paramagnetic state for no applied field, will vary as $(T^{4/3})^2$. This may provide an explanation for observed temperature dependence of the susceptibility in $Ni_3Ga$. We can calculate this more quantitatively within this extended version of the SCR model. Figure 5 shows the experimental data compared with calculations based on (i) the initial equation of state given by equation (1) and (ii) the extended version of the equation of state given by equation (6) in which the mode coupling parameter $b$ is close to zero. We should point out that these calculated results depend on the range over which the fit to the low-temperature equation of state is performed and so the exact magnitude of the predictions should not be taken too seriously. However, it is clear from figure 5 that the case in which the mode coupling parameter, $b$, is close to zero gives a much better explanation of the data. In particular this explains the anomalous temperature dependence of the susceptibility. This suggests that the mode coupling parameter, $b$, is close to zero indicating that the system is reasonably close to a tri-critical point (the point where $A = 0$ and $B = 0$). In addition, the fact that the higher-temperature Arrott plots become linear is also qualitatively consistent as at higher temperatures the $B$ parameter becomes positive (due to the temperature dependence given in equation (8)) and so the $bM^2$ term begins to dominate over the higher order $gM^4$ term.

## 5. Conclusion

The main conclusion of this letter is that the temperature dependence of the susceptibility in $Ni_3Ga$ is affected by the tri-critical point and is consistent with its presence.

Previously, the temperature dependence of the inverse susceptibility in $Ni_3Ga$ was thought to contradict the SCR model as the temperature dependence was $T^2$ rather than the expected $T^{4/3}$. However, it has been shown here that this behavior can be explained by a SCR approach if the proximity of the system to a quantum tri-critical point is taken into account. Although we have not directly taken into account the non-analytic corrections to the SCR model we are in no way dismissing their importance. By using higher order terms in the expansion for the equation of state we have sought to capture some of the physics that these non-analytic corrections contain. While using an analytic expansion for a non-analytic function is clearly ultimately invalid we believe it is still of value as it enables a relatively simple comparison between theory and experiment.

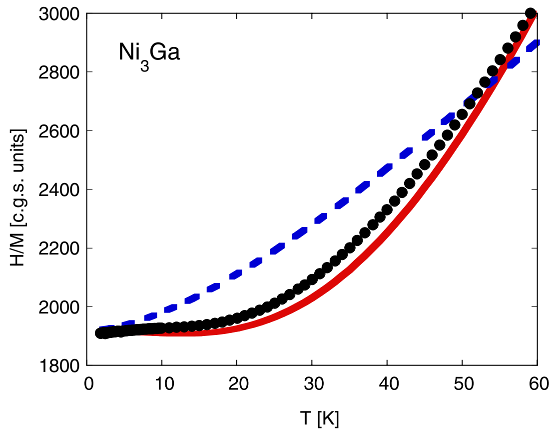


**Figure 5.** The inverse volume susceptibility against temperature for $Ni_3Ga$. The experimental data, shown as black circles, are compared with the susceptibility calculated using equation (2) (blue dotted line) with $a = 1920$ and $b = 0.38$ $G^{-2}$ (with $b$ taken from previous measurements at higher temperature [23]) and that calculated using equation (7) (red line) with $a = 1920$, $b = -0.1$ $G^{-2}$, and $g = 1.5 \times 10^{-4}$ $G^{-4}$ (where these parameters are taken from a fit of the form $H/M = a + bM^2 + gM^4$ to the data given in figure 4). In both the calculations the parameters $c \approx 1.0 \times 10^5$ Å$^2$ and $\hbar\gamma \approx 3.0$ $\mu$eV Å were estimated from inelastic neutron scattering measurements.

Our indirect evidence for the tri-critical point is consistent with what has been seen in several other systems on the border or magnetism [24, 25]. As we have said, the prevalence of such a point may result from the non-analytic corrections to the magnetic equation of state in the SCR model mentioned above. In addition, band structure effects such as proximity to a van-Hove singularity may also add additional structure to the equation of state and the momentum-dependent susceptibility (Lindard function) [26, 27].

The key new result is that we have demonstrated the existence of different quantum critical exponents which result not from a quantum critical point but from a quantum tri-critical point. We therefore suggest that this quantum tri-critical point may be a key feature more generally in quantum critical systems and may point toward an explanation of the many emergent non-Fermi liquid phases that have been observed.

## Acknowledgments

I thank Gil Lonzarich for suggesting this work and for his support under EPSRC grants EP/C009487/1 and EP/C511778/1. I also thank M. Ellerby, S S Saxena, S E Rowley and A Kusmartseva for help with the measurements and N R Bernhoeft and S M Hayden for the samples. I acknowledge financial support from EPSRC and St Catharine's College, Cambridge.

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