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Pressure Studies on the Magnetic Phase Diagram of $\mathrm{Cr}_{.995}\mathrm{Rh}_{.005}$

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Abstract

The pressure dependence of antiferromagnetic ordering in $\mathrm{Cr}_{.995}\mathrm{Rh}_{.005}$ alloy has been investigated. Anomalies in resistivity and thermopower near Neel temperature have been used to delineate the magnetic phase boundary in these systems. The important finding of this study is that the commensurate - incommensurate phase boundary in the P-T plane can be delineated using the anomalies in thermopower (TEP) across the transition. These new results find a simple explanation based on a model for electron transport applicable to chromium alloys developed by Trego and Mackintosh.[1]

1 Introduction

It is well known that chromium and its alloys exhibit a variety of magnetic phase transitions associated with the nesting of the Fermi surface. Pure chromium is an itinerant antiferromagnet described as an incommensurate spin density wave below the Neel temperature (311 K). This unique type of antiferromagnetic ordering has been explained by Lomer[2] and Overhauser[3] as due to the attractive Coulomb interaction between electron and hole pockets near the Fermi surface which have approximately the same shape (octahedral) and size. It is clear that such an attractive interaction leads to an energy gap at the Fermi surface. This feature gives rise to well defined anomalies in electrical resistivity and thermoelectric power.[1] Alloying studies have established that there is a correlation between the electron to atom ratio (e/a) and the Neel temperature (T_N) and also the type of antiferromagnetic ordering.[4] Experimentally it has been observed that for $e/a < 6$, T_N is lowered on alloying and the spin density wave (SDW) remains incommensurate. However alloying with an element whose $e/a > 6$ leads to a marked increase in T_N and more importantly at a critical concentration the magnetic order changes to a commensurate structure. The current experimental and theoretical situation in SDW antiferromagnetism of chromium alloys has been excellently reviewed in a recent article by Fawcett et al.[5]

The effect of pressure on different magnetic transitions like commensurate - paramagnetic (C-P), incommensurate - paramagnetic (I-P) and incommensurate - commensurate (I-C) in chromium alloys is of fundamental interest. The striking similarity in the phase diagrams in the pressure - temperature plane and the composition - temperature plane in these systems has led to the general view that increasing pressure is equivalent to decreasing the electron concentration in Cr SDW alloy systems.[6,7] However Griessen and Fawcett[8] and Fawcett[9] have pointed out that this empirical correspondence does not mean that the effect of pressure and that of changing the electron concentration are equivalent.

In this paper we present new results on the $Cr_{0.995}Rh_{.005}$ alloy system where both electrical resistivity and TEP are used to track the different magnetic transitions in the P-T plane. A significant aspect of the present experimental study is that TEP is a convenient tool to probe the magnetic phase boundaries especially in the high pressure region where the characteristic resistivity anomaly vanishes. Isothermal runs of TEP vs pressure in this system have provided for the *first time* direct evidence for the commensurate - incommensurate magnetic phase transition whose phase boundary cannot be located by other transport property measurements. We also propose that the high pressure behaviour of resistivity and TEP in this system can be understood on the basis of a simple model developed by Trego and Mackintosh.[1]

2 Experimental details

Chromium alloys were prepared by arc melting and homogenised at 1200^0C and then furnace cooled. High pressure measurements on these annealed samples were carried out in the teflon thermopower cell[10] in a conventional piston-cylinder apparatus. Silicone oil of viscosity grade 150 cst was used as a pressure transmitting medium. *AC resistivity* measurements were carried out using the techniques developed by the authors described elsewhere.[11,12] A PC based data acquisition system with software control of mean temperature, temperature gradient across the sample, data acquisition and analysis developed by the authors was used for high resolution TEP measurements.[13] It would suffice here to mention that the ease with which the parameters of the three mode (PID) temperature controller can be changed in the software has greatly contributed to the tight temperature control achieved in the teflon thermopower cell. Further real-time digital signal processing has led to a significant improvement in the signal to noise ratio. In this system temperature stability of the order of ±0.02^{0}C upto 300^{0}C has been achieved. The resolution in TEP measurement is around 0.01 μV/^{0}C. Resistivity changes of the order of one part in a thousand can be easily detected in this system.

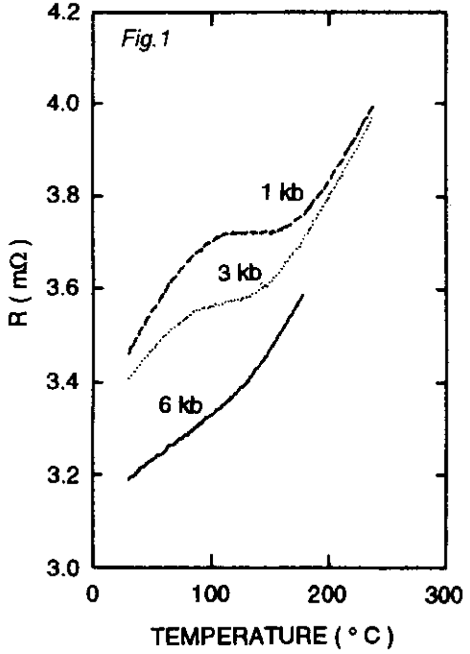
3 Results

3.1 Resistivity studies across C-P phase transition

Fig 1 gives the resistance vs temperature behaviour of this alloy in the low pressure region upto 6 kbar. The resistivity anomaly near Neel temperature (T_N) manifests as a weak minimum for this alloy. This behaviour should be contrasted with the hump observed in higher concentration alloy.[14] It must be mentioned that for the Cr-Rh system the critical concentration is around 0.11 atomic % of Rh[15] beyond which the SDW becomes commensurate with a marked increase in the magnitude of T_N. It is clear that in the alloy under study viz., $Cr_{0.995}Rh_{.005}$, the SDW is commensurate at ambient pressure and temperature conditions. The anomaly near 175^{0}C (T_N) at 1 kbar is thus associated with the commensurate - paramagnetic phase transition. Pressure has a marked effect on the nature of the resistivity anomaly accompanied by a strong depression in the value of T_N. In the isobar at 6 kbar the C-P transition is observed only as a change of slope around 120^{0}C. Fig 2 presents several isobars in the high pressure region. We note that the resistivity anomaly almost vanishes in the pressure region beyond 12 kbar and makes it impossible to track the magnetic transition at higher pressures.

3.2 TEP studies across C-P & I-P phase transition

The isobars of absolute TEP vs temperature upto 10 kbar pressure are given in Fig 3. As viewed from the high temperature side, TEP exhibits a marked increase in the magnitude as the sample is cooled below T_N (around 200^{0}C at 0.5 kbar) reaching a maximum at a temperature lower than T_N. This large TEP anomaly (≈ 9 μV/^{0}C) is in sharp contrast to the rather weak resistivity anomaly shown in Fig 1. We note that the position of the peak in the TEP vs temperature plot shifts to lower temperatures with increase in pressure. The shift of T_N with pressure associated with the C-P transition is rather large $\approx$ -5^{0}C/kbar and can be easily evaluated using TEP anomaly as a marker. Fig 4 shows the isobars of TEP where both the C-P and I-P magnetic transitions exhibit characteristic anomalies. The 12 kbar isobar is associated with the C-P transition. There is a marked change in the nature of the TEP anomaly at higher pressures as reflected in the 15 kbar curve. Firstly the position of the peak in the TEP vs temperature plot has shifted to higher temperatures ($\approx 80^0$C) whereas one would have expected this peak to occur at a temperature lower than 40^0C had the commensurate phase been stable in this pressure region. Secondly the magnitude of the anomaly has reduced considerably. This clearly indicates that at a critical pressure between 12 and 15 kbar the commensurate phase changes over to an incommensurate antiferromagnetic SDW. The Neel temperature here is associated with the I-P transition. Fig 5 presents the TEP data on an expanded scale for 20 kbar and 23 kbar pressure. The scatter in the data is $\approx \pm 0.01\ \mu$V/^{0}C. It is important to note that the characteristic anomaly near T_N is still observable. The shift of T_N



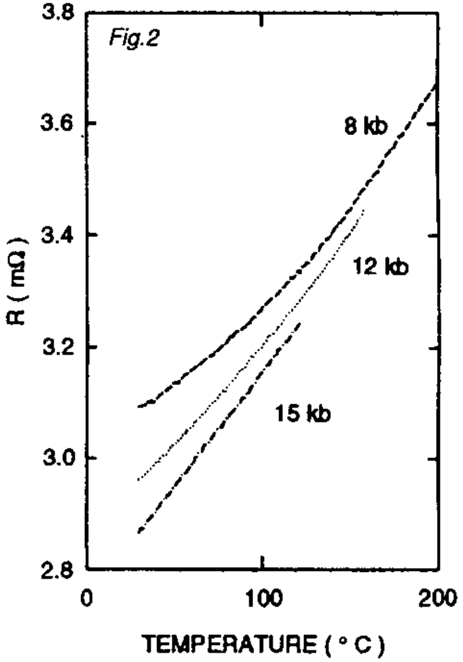
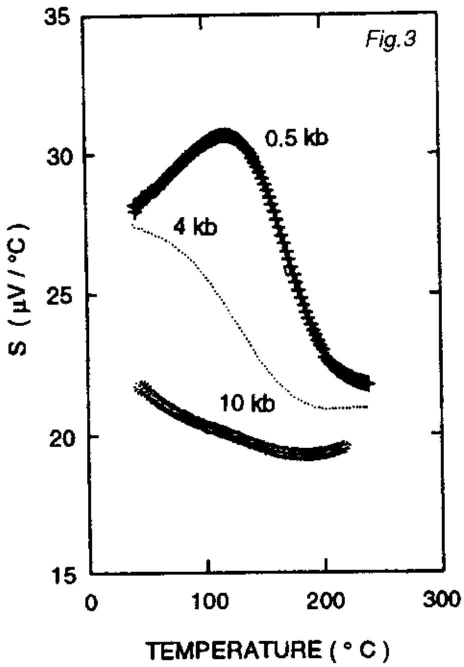


Figure 1: *Resistance vs temperature at lower pressures.*

Figure 2: *Higher pressure isobars.*

with pressure is small ≈ - 2^0C/kbar which is typical of the slope of the I-P phase boundary. The present experimental studies clearly indicate that TEP is a potent tool to track the magnetic phase boundaries at pressures even beyond 25 kbar.



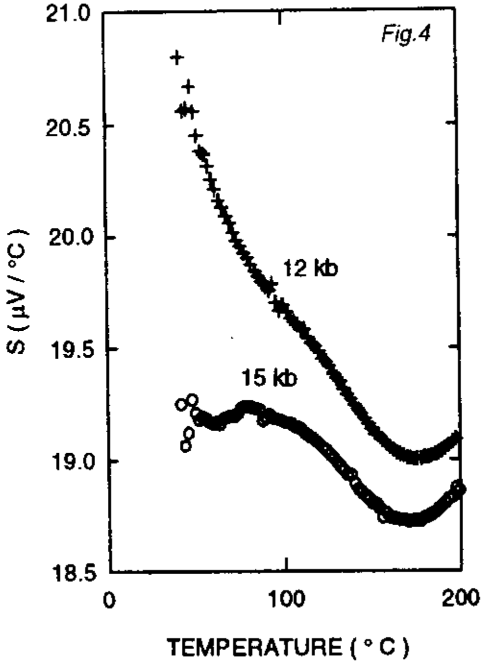


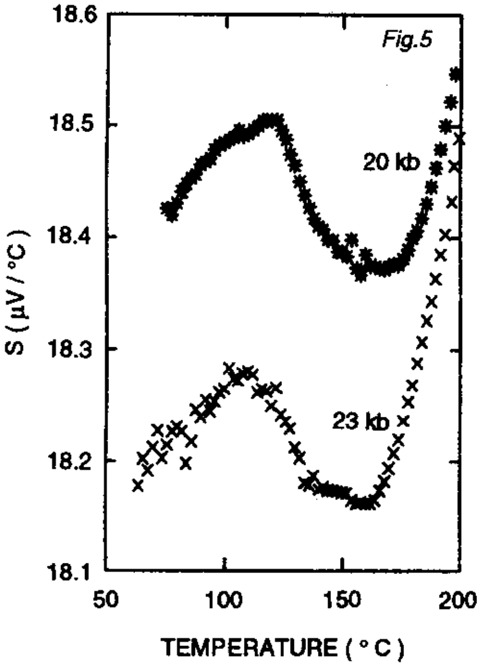
Figure 3: *TEP vs temperature in the low pressure region.*

Figure 4: *Change over from C-P to I-P transition at higher pressures.*

3.3 TEP studies across C-I phase transition

The C-I transition is nearly always continuous except in systems like CrMn and CrFe. Resistivity does not exhibit any anomaly across this phase transition. We have tried to locate this phase boundary in the P-T plane through careful measurements of TEP. Fig 6 presents TEP data as a function of pressure for two isothermal runs at 50^0C and at 170^0C. It is worth mentioning that the mean temperature of the sample was held constant to within ± 0.02^0C during pressurisation. We observe distinct anomalies near 9.5 kbar for the 50^0C isotherm and near 10.75 kbar for the

170^{0}C isotherm. We believe that these anomalies are associated with the C-I phase transition. The slope of the C-I phase boundary is positive and large ≈ 100^{0}C/kbar. A magnetic phase diagram delineating C-P, I-P and C-I phase boundaries in the P-T plane is given in Fig 7. The main feature of this diagram is the existence of a **triple point** with co-ordinates 10.5 kbar and 170^{0}C where all the three phases co-exist.



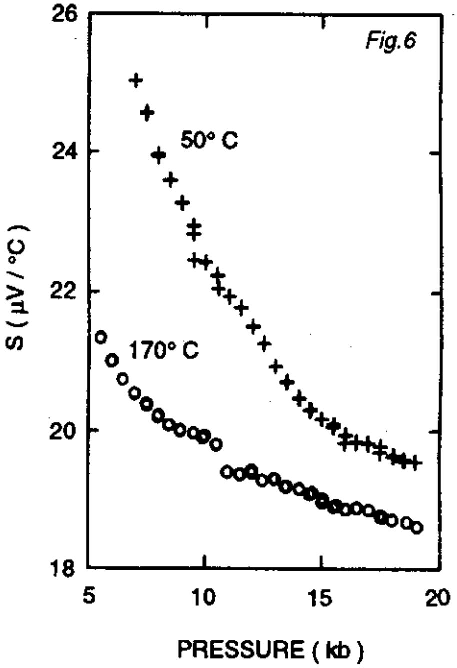


Figure 5: *I-P transition as studied through TEP.*

Figure 6: *TEP anomaly near C-I phase boundary.*

4 Discussion

It is well known that the itinerant antiferromagnetism of Cr is related to its peculiar band structure.[2,3] The attractive Coulomb interaction between electrons and holes in two different sheets of Fermi surface separated by a wave vector Q in the (100) direction leads to an energy gap in the single - particle spectrum.[16] Essentially a part of the Fermi surface is wiped out at the Neel temperature due to this attractive

Coulomb interaction. It is this feature which gives rise to well defined anomalies in the electronic transport properties. The present experimental results, especially those on TEP can be qualitatively understood on the basis of a model developed by Trego and Mackintosh.[1] Although the arguments presented in this model are applicable to the incommensurate phase, qualitatively the same results hold good for the commensurate phase also. It is well known that the diffusion part of TEP is related to the energy derivative of the density of states and the carrier relaxation time (τ_{ph}) evaluated at the Fermi energy. In this model although the density of states undergoes a marked decrease at the antiferromagnetic transition, its energy derivative is not expected to change drastically. The major contribution to TEP is then due to the term $[d\tau_{ph}/dE]_{E=E_F}$. The electron - phonon scattering term which determines τ_{ph} is drastically affected due to the energy gap 2Δ right at the Fermi energy. Since the number of final states into which an electron at the Fermi surface can be scattered

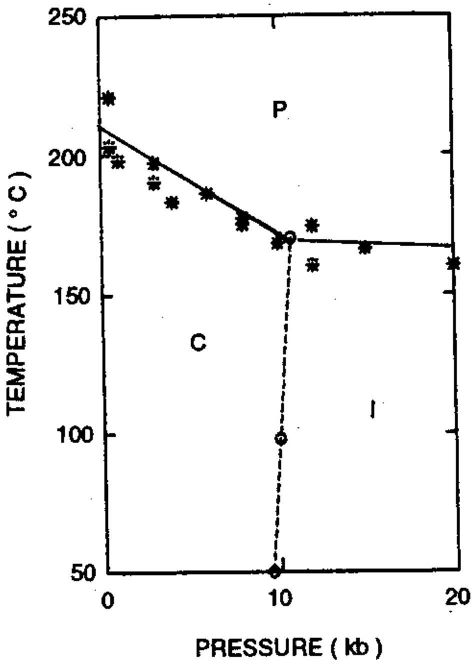


Figure 7: *Magnetic phase diagram.*

by a phonon is highly reduced, this has the effect of increasing τ_{ph}. Assuming that the phonon spectrum may be represented by the Debye type, these authors evaluated the energy derivative of τ_{ph} for various values of Δ. It turns out that this derivative is negative and thus makes a positive contribution to TEP. The energy gap in the antiferromagnetic phase is also temperature dependent becoming zero at T_N. Thus, as the gap 2Δ increases from a value smaller than E_{ph} (the characteristic energy of the phonon) to a value higher than E_{ph}, this derivative term reaches a negative maximum when $\Delta(T) \approx E_{ph}$. This leads to a positive maximum in the behaviour of TEP at a temperature below T_N. Experimentally this type of variation is observed in pure Cr and also in alloys. The hump in TEP vs temperature plot is however more marked in the commensurate phase as compared to the incommensurate phase[1].

The TEP vs temperature data at different pressures given in Figs 3-5 follow the pattern to be expected from the above model. The energy gap $2\Delta(T)$ is however pressure dependent, decreasing with pressure at different rates in the commensurate and incommensurate phases. The magnitude of the hump in the TEP vs temperature plot (Fig 3), characteristic of the commensurate phase with $\Delta \approx 0.4$eV, decreases with increase in pressure and the position of the peak shifts to lower temperatures. This is in conformity with the conclusions of the above model where the peak is identified by the condition $\Delta(P) \approx E_{ph}$. We have also used this criterion to suggest that the 15 kbar isotherm (Fig 4) is associated with the I-P transition by noting that the magnitude of the hump in the TEP plot is much smaller which is typical of an incommensurate phase ($\Delta \approx 0.1$eV) [Fig 5].

The direct observation of the C-I phase boundary through high resolution TEP measurements (Fig 6) is an interesting aspect of the present experimental studies. It appears from these data that the pressure coefficient of the energy gap in the commensurate phase is larger than that in the incommensurate phase.

The blurring of the resistivity anomaly at higher pressures (viz., 15 kbar isotherm in Fig 2) can also be accounted for by noting that $2\,\Delta(P) \ll E_{ph}$, so that the effect of the energy gap is not seen at all.

In summary, we note that studies of TEP in the P-T plane are valuable in delineating the magnetic phase boundaries and in the understanding of the pressure effects on the electronic structure of Cr based alloys.

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