Giant positive magnetoresistance and field-induced metal insulator transition in Cr$_2$NiGa

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Abstract
We report the magneto-transport properties of the newly synthesized Heusler compound Cr$_2$NiGa which crystallizes in a disordered cubic B2 structure belonging to the $Pm\bar{3}m$ space group. The sample is found to be paramagnetic down to 2 K with metallic characteristics. On application of a magnetic field, a significantly large increase in resistivity is observed which corresponds to magnetoresistance as high as 112% at 150 kOe of field at the lowest temperature. Most remarkably, the sample shows a negative temperature coefficient of resistivity below about 50 K under the application of field $\geq 80$ kOe, signifying a field-induced metal to ‘insulating’ transition. The observed magnetoresistance follows Kohler’s rule below 20 K indicating the validity of the semiclassical model of electronic transport in metals with a single relaxation time. A multi-band model for electronic transport, originally proposed for semimetals, is found to be appropriate to describe the magneto-transport behavior of the sample.

Keywords: Heusler alloy, magnetoresistance, metal insulator transition

(Some figures may appear in colour only in the online journal)

1. Introduction
The intermetallic Heusler [1] system of alloys continue to be the forefront of active research since its discovery about a century ago. Heusler alloys reveal wealth of fascinating properties including half-metallicity [2–4], magnetic shape memory effects [5, 6], unconventional superconductivity [7] and so on. In general, they represent a class of ternary intermetallic compounds having the general formula X$_2$YZ where X and Y are transition metals whereas Z is a non-magnetic $sp$ element. Ideally, Heusler compounds crystallize in the Cu$_2$MnAl-type ordered L2$_1$ structure consisting of eight stacked body-centered-cubic lattices. The most common disordered structure arising from L2$_1$ is B2 (CsCl-type structure) which occurs due to random occupancy of Y and Z atoms. In this case, Y and Z sites become equivalent with the lowering of crystal symmetry (space group no. 221: $Pm\bar{3}m$) [8].

Recently, Cr-based stoichiometric (Cr$_2$YZ) full Heusler compounds have become an interesting topic of investigation. Particularly, systems having 24 valance electrons are promising candidates for being half-metallic compounds [9–11]. Among them, Cr$_2$CoGa (crystallizes in the XA inverse Heusler structure) has been predicted to be a half-metallic fully compensated ferrimagnet where the moments from two Cr sites (at X and Z positions) compensate each other resulting in a zero macroscopic moment [9]. Here we have studied the nearest sibling of the compound, namely, Cr$_2$NiGa which has 25 valance electrons. Up until now there has been no report of the formation of the compound experimentally, nor is there any report on the electronic or magnetic properties based on theoretical calculations. In contrast to the Co-counterpart,
Cr$_2$NiGa is found to be paramagnetic (PM) with rather unusual magneto-transport properties at low temperature.

2. Experimental details

Polycrystalline samples of Cr$_2$NiGa were prepared by standard arc melting technique. Next, the arc melted ingot was vacuum sealed in a quartz capsule and annealed at 900 °C for 48 h followed by normal furnace cooling down to room temperature. In order to get confirmation about the stoichiometry of the alloy, we performed energy-dispersive x-ray spectroscopy (EDS) by scanning different parts (effective scanning area of $0.5 \times 0.5 \text{ mm}^2$) of the sample using JEOL JSM 6700F. The structure illustrates the 3D-network in disordered cubic Heusler phase of Cr$_2$NiGa having disorder between Ni and Ga.

3. Results

3.1. Powder x-ray diffraction

Figure 1 depicts powder XRD pattern of the studied sample measured at room temperature. Observation of some strong Bragg reflections in the XRD pattern initially indicate a simple body centered cubic (bcc) structure. Rietveld refinement of the pattern confirms the formation of CsCl-type B2 structure, where Ni and Ga atoms randomly occupy Y and Z sites. We can simply rule out the formation of ordered L2$_1$ or XA structures from the absence of a superlattice line around $2\theta \sim 26^\circ$. Best fitting is obtained by considering partial occupancies of Ni and Ga for the equivalent Y/Z position of X$_2YZ$ Heusler structures with a standard deviation $\sigma = 1.5046$. For the refinement, Ni occupancy was 0.57 whereas Ga has 0.43 occupancy and calculated cubic lattice parameter is found to be 2.8967 Å.

3.2. Magnetization

We present the temperature ($T$) variation of magnetic susceptibility ($\chi = M/H$, where $M$ is the measured magnetization and $H$ is the applied magnetic field) in the presence of $H = 1$ kOe from 380 to 2 K in figure 2. It is evident that $\chi$ increases monotonically with decreasing $T$ and we do not observe any signature of magnetic anomaly. The $\chi(T)$ data can be well fitted with the Curie–Weiss law (solid line in the plot) where an additional temperature independent susceptibility term is considered ($\chi = C(T - \theta) + \chi_0$). The effective PM moment per formula unit and the PM Curie temperature of the sample are found to be $\mu_{eff} = 0.71 \mu_B$ and $\theta = -6.4$ K, while the value of $\chi_0 (\sim 10^{-5} \text{ emu mol}^{-1})$ is found to be small but positive. The presence of small but finite $\chi_0$ is possibly due to the spin fluctuation enhanced Pauli paramagnetism in this itinerant electron system [13].

The PM nature of the sample is further clarified from the isothermal magnetization measurement. The inset of figure 2 represents $M(H)$ curves measured at $T = 2, 25, 50$ and 300 K for 0 to 50 kOe field cycling. The 2 K isotherm shows clear signature of curvature, while the other isotherms are found to be linear in nature. We argue that the curvature is the manifestation of inherent non-linear nature of the $M - H$ curves for free PM moments ($\mu$) which is particularly visible at low $T$ and high $H (k_B T / \mu H \ll 1)$. We used the classical Langevin function for PM material to fit the data with $M(x) = M_0 [\coth(x) - 1/x]$. 

\[ \chi = \frac{\chi_0}{1 - T/\theta} \]

where $\chi = M/H$, $M$ is the measured magnetization, $H$ is the applied magnetic field, $\chi_0$ is the temperature independent susceptibility term, $\theta$ is the Curie temperature, and $T$ is the absolute temperature. The fitted curves to the experimental data.
with \( x = \mu H/k_B T \). The fitting is shown in the inset of figure 2 by the solid line for 2 K isotherm. The good fit once again indicates the PM nature of the solid. Evidently, the sample has a quite low value of \( \mu (0.0214 \text{ B} \mu_\text{B} \text{ f.u.}^{-1} \) at 2 K for \( H = 50 \text{ kOe} \), which is presumably related to the itinerant nature of the Cr or Ni moments.

3.3. Magneto transport

The most fascinating observation in the present work is obtained from the magneto-transport measurement on the sample. The \( T \) variation of \( \rho \) during heating in the presence of \( H = 0, 20, 50, 80, 90, 120 \) and 150 kOe (figure 3(a)). For all the measurements, the magnetic field was in the transverse direction with respect to the current. The zero-field \( \rho(T) \) curve shows pure metallic nature as evident from the continuous drop in \( \rho \) with a decrease in \( T \). The residual resistivity ratio (RRR = \( \rho(300 \text{K})/\rho(4 \text{ K}) \)) is found to be 5 with the value of \( \rho(4 \text{ K}) = 5 \mu\Omega \text{ cm} \), signifying a good metallic character of the sample. All the \( \rho(T) \) curves measured in the field overlapped with the \( H = 0 \) curve in the high \( T \) region (above about 135 K), whereas, deviation between them is visible at low temperatures. \( \rho(T) \) becomes flat below about 50 K for \( H = 20 \) and 50 kOe. However, for \( H = 80 \text{ kOe} \) and above, \( \rho(T) \) show an upward turn around 50 K and increases (\( d\rho/dT < 0 \)) down to the lowest temperature. Such an upturn indicates that the system undergoes a field-driven transition from a pure metallic to a state having ‘insulating’ characteristics. For clear visualization, low \( T \) parts of \( \rho \) for \( H = 80, 90 \), 120 and 150 kOe are shown in the upper inset of figure 3(a).

Large increment in \( \rho \) with \( H \) at low \( T \) signifies large positive magnetoresistance (MR) in the sample. Thermal variations of calculated MR \( [= (\rho_H - \rho_0)/\rho_0 = \Delta\rho/\rho_0] \) from the different iso-field \( (H = 20, 50 \) and 150 kOe) \( \rho(T) \) curves are plotted in the main panel of figure 3(b). Here, \( \rho_0 \) and \( \rho_H \) have resistivity in the zero field and in the presence of \( H \). We observe a huge 112\% of positive MR at 8 K in \( H = 150 \text{ kOe} \). The MR is also reasonably high at lower \( H \), for example, MR is about 50\% at low temperature \( (\sim 7 \text{ K}) \) for \( H = 50 \text{ kOe} \). With increasing \( T \), magnitude of MR decreases and tends to vanish as \( T \) approaches 135 K.

We have carried out isothermal \( H \) variation of \( \rho \) at different constant \( T \). Since the significant field effect is only observed below 135 K, we have restricted our measurement up to this temperature. For this purpose, \( \rho \) was measured at the selected constant \( T \) by cycling \( H \) between \( \pm 140 \text{ kOe} \) and the corresponding MR was calculated (see inset of figure 4). For the sake of clarity, only positive \( H \) quadrant is presented. Apparently, all the measured isothersms show a quasi-linear nature with varying slope depending upon \( T \). The 8 K isotherm shows MR of magnitude 112\%, which is in accordance with the iso-field \( \rho(T) \) data. At high-\( T \), MR is quite small and for example, it is about 8\% at 130 K.

Positive MR in a metal is often attributed to the curving of electron trajectory due to the application of \( H \) which effectively reduces the electronic mean free path (\( \lambda \)) [14, 15]. This model gives positive MR only when there are more than one type of carriers and a quadratic dependence of MR on \( H \) is obtained. For the present sample, MR is quasi-linear contrary to the usual observation. We have fitted the field dependence of MR data by the relation \( \Delta\rho/\rho_0 = \alpha H^n \) where \( \alpha \) is the coefficient of power law. The values of \( n \) are found to be close to unity and lie between 0.89 to 1.23 which is well below the value 2 expected for a PM or non-magnetic metal.

We have carefully looked at the low-\( T \) part of the data by plotting \( \rho \) as a function of \( T \). \( \rho \) does not obey a simple \( T^2 \) dependence in accordance with the Fermi liquid behavior. However, there are numerous reports in the literature, where the low-\( T \) part of a Fermi metal can be described by a slightly modified law containing \( T^3 \) term resulting \( \rho(T) = \alpha_0 + \alpha_2 T^2 + \alpha_4 T^4 \) below 50 K. (b) Shows \( T \) variation of iso-field MR for different constant \( H \). Inset shows the variation of \( \lambda \) and \( \Delta\rho/\rho_0 \) (for 150 kOe) with temperature.

![Figure 3](image-url)
Interestingly, thermal variation curves for both the parameters $\Delta \rho / \rho_0$ and $\rho(H) - \rho(0)$ at selective temperatures. Red lines are the fitted curves to the isotherms. Inset shows isothermal variation of MR measured behavior can follow an empirical relation: $\Delta \rho / \rho_0 = F(H / \rho_0)$, where $F$ is a function characterized by the inherent electronic structure and external geometry of the metal [14, 17]. Here, $\rho_0$ can be expressed as $\rho_0 = m^* n e^2 \tau$ for free electrons with $\tau$ being the characteristic relaxation rate of scattering. The effective electronic mass, volume density of conduction electrons and electronic charge are denoted by $m^*$, $n$ and $e$, respectively. In the light of the semiclassical theory of transport, Kohler’s rule should only be valid for a metal having a single type of charge carrier along with the equal $\tau$ at all the points on the Fermi surface. Accordingly, $\Delta \rho / \rho_0$ plotted as a function of $F(H / \rho_0)$ should be $T$ independent. Since our data indicates quasi-linear variation of MR with $H$, we have chosen $F(H / \rho_0)$ to be equal to $H / \rho_0$. Interestingly, all the MR curves in the Kohler’s plot up to 20 K collapse on each other signifying the validity of the rule. However, above 20 K we see marked deviation from Kohler’s rule as the curves are found to be strongly $T$ dependent. Such violation of Kohler’s rule at high-$T$ is likely to be related to the enhanced electron–phonon scattering which breaks the uniqueness of $\tau$.

We have calculated $\lambda$ of the conduction electrons on the framework of the Drude–Sommerfeld model where

$$\lambda = \frac{\tau v_F}{\hbar} = \frac{\hbar}{n e^2} (3 \pi^2 n)^{1/3}.$$  

Here, $v_F$ is the Fermi velocity [18]. The value of $\lambda$ is found to be around 40 Å at 5 K, which indicates that $\lambda$ is large enough (it is much larger than the interatomic spacing) so that the system can be described by the semiclassical model. We have plotted thermal variation of $\Delta \rho / \rho_0 (H = 150$ kOe) along with the calculated $\lambda(T, H = 0)$ in the inset of figure 3(b). Interestingly, thermal variation curves for both the parameters are quite similar signifying close correspondence between mean free path of scattering (and as a result $\tau$) and the magnitude of MR. Such correspondence as well as the validity of Kohler’s law (at least for $T \ll 20$ K) imply that the observed magneto-transport behavior of Cr$_2$NiGa can be well accommodated within the semiclassical model for electronic conduction in metals with a single $\tau$ [15, 19].

4. Discussion

The observed MR in Cr$_2$NiGa is substantially high particularly for a polycrystalline intermetallic sample with static defects and disorders (residual resistivity is about 5 $\mu\Omega$ cm). The present sample does not show any signature of a long range magnetic ordering down to 2 K. Therefore, the observed large positive MR is unlikely to have a magnetic origin. Such conclusion is also supported by the fact that our MR data corroborates well with the free electron model. Even if there is some magnetic correlation present in the sample, it is not playing any role towards the magneto-transport properties of the system.

There are few examples of large positive MR in bulk intermetallic systems [20–22], and in many cases, MR is found to be uncorrelated with the ordered magnetic state of the system. The most important observation in the present study is the remarkable metal to ‘insulator’ transition in high applied fields. Possibly, the trend for a semiconducting nature starts at a lower field (the positive MR is an outcome of that), and eventually leads to a state with negative $T$ coefficient of $\rho$. The semiconducting behaviour of the sample can have several possible origins, such as weak localization, the Kondo effect or due to the opening up of a soft gap at the Fermi level. We can rule out localization or the Kondo process, as $H$ should drive the system to have lower $\rho$ causing negative MR. The $\rho$ versus $T$ plot under $H = 150$ kOe also does not follow log $T$ variation below the $\rho$ minimum. We also failed to observe any activated behavior ($\rho \sim \exp(-1/T)^{\beta}$, $\beta$ is a generalized parameter $\leq 1$) that fits well with an electronic gap. A gap in the Fermi level would also expect to alter $m^* n e^2$, which in its turn should violate Kohler’s rule [17].

Interestingly, similar positive MR and metal-insulator transition had been reported for several semimetals [23–25] including Bi, Sb and graphite [26, 27] (reported mostly on high mobility single crystalline samples). Surprisingly, the magneto-transport properties of these semimetals qualitatively resemble quite well with the presently studied Cr$_2$NiGa compound. In case of the semimetals mentioned above, the insulating state develops at a rather low magnetic field (can be as low as 1 kOe) [25] as compared to one hundred of kOe in the present Heusler system. Nevertheless, the field induced ‘insulating’ part shows similar $T$ variation with identical saturating behavior at the lowest $T$. Among many plausible explanations, a simple multi-band model has been found to be quite successful in explaining such metal to insulator transition as well as a positive MR. In this model, total $\rho(T)$ is found to contain an $H$ independent metallic part and an $H$ dependent ‘insulating’ part which vanishes for $H = 0$ [25]. The ‘insulating’ part dominates at lower $T$, surpassing the metallic contribution and thereby turning overall $\rho$ to increase with decreasing $T$. Although, semi-metallicity in full Heusler alloys are not known, similar multi-band models can be generalized.
and found useful for explaining these anomalous magneto-
transport properties of the studied compound.

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