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Cite as: AIP Advances **6**, 055815 (2016); https://doi.org/10.1063/1.4943609
Submitted: 06 November 2015 . Accepted: 28 December 2015 . Published Online: 04 March 2016

Sk Mohammad Yasin, Ritwik Saha, V. Srinivas, S. Kasiviswanathan, and A. K. Nigam

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Giant magnetoresistance in the cluster glass regime of Co-Ga alloys

Sk Mohammad Yasin,[1] Ritwik Saha,[2] V. Srinivas,[1,a] S. Kasiviswanathan,[1] and A. K. Nigam[2]
[1]*Department of Physics, Indian Institute of Technology Madras, Chennai 600 036, India*
[2]*Department of Condensed Matter Physics and Materials Science, Tata Institute of Fundamental Research, Mumbai 400 005, India*

(Presented 14 January 2016; received 6 November 2015; accepted 28 December 2015; published online 4 March 2016)

A detailed study of low temperature electrical transport properties of Co_xGa_{100-x} (x = 54, 55.5, 57) alloy has been carried out. The origin of the resistivity anomalies and correlation between magnetic and electrical transport properties are identified through an elaborate analysis. The weak localization and enhanced electron-electron interaction effects partially support the electrical transport properties of the system. Further, the observed magnetoresistance can be well represented by localized model along with quantum corrections. The low temperature magnetoresistance value near critical composition is comparable to that reported in giant magnetoresistance materials. [http://dx.doi.org/10.1063/1.4943609]

I. INTRODUCTION

Materials with weak magnetic properties and narrow energy band gap in their electronic band structure appear to generate unusual properties yet useful for various applications, such as, giant magnetoresistance (GMR), large thermopower and giant Hall effects. Therefore, a search for materials exhibiting these characteristic features was launched to exploit them for the development of novel and efficient electronic devices. Transition metal aluminides, silicides and gallides with composition close to the equiatomic composition seem to be quite promising.[1–4] From the electronic band structure calculations on equiatomic Co-Ga, a pseudogap near the Fermi-level in the DOS spectrum has been predicted.[4] These results were supported by the experimental findings, where non-magnetic behaviour and higher electrical resistivity values were reported.[4–6] These studies also revealed that the samples stabilize in same crystal structure with a significant change in magnetic and electrical properties. This lead to a belief that Co antisite (AS) defect is associated with an effective moment of $\sim 5\mu_B$.[7,8] Subsequently more complex models have been proposed ascribing magnetic behavior only to clusters of AS atoms while a single AS defect carries no moment.[9,10] Further, the magnetic phase diagram suggests the presence of a range of magnetic phases, such as, cluster spin glass to ferromagnetic (FM) state with small variations in composition.[11] On the other hand, earlier reports on electrical resistivity present a qualitative study over a wide range of temperature. A significant change in resistivity and temperature coefficient of resistivity (TCR) values especially in low temperature regime has been reported.[4–6] These properties are also sensitive to the process conditions which suggests that both band structure and localized moments play equally important role in altering the physical properties. From these studies it is not clear whether such variations are due to changes in the DOS or scattering of electrons from localized moments. We believe the electrical transport studies under magnetic field along with composition variations (below critical composition) can throw some light on the transport anomalies. Further, a detailed

[a]Corresponding author. E-mail address: veeturi@iitm.ac.in

study on the low temperature magnetotransport has not been reported on the alloys close to critical concentration. In this work we present a detailed low temperature electrical transport study on Co_xGa_{100-x}(x = 54-57) system to unravel the issues related to low temperature electrical transport behaviour.

II. EXPERIMENTAL DETAILS

The Co_xGa_{100-x}(x = 54-57) alloy ingots were prepared by arc melting high purity constituents followed by annealing at 873 K for 72 hrs and then allowed to furnace cool. The structure and chemical compositions were confirmed through powder x-ray diffraction (XRD) and energy dispersive spectroscopy (EDS) measurements (Quanta 200 SEM) respectively. Electrical transport measurements were carried out in physical properties measurement system (PPMS) in the temperature range 2-300 K. The electrical transport measurements were carried out with standard four probe method on a sample with dimensions 10 mm × 5 mm × 1 mm. The MR measurements were carried out by applying magnetic field (H = 0-90 kOe) perpendicular to the transport current.

III. RESULTS AND DISCUSSION

The temperature variation of normalized electrical resistivity, $\rho(T)/\rho(2K)$, for x = 54-57 compositions is shown in Fig. 1. From the figure one can notice following features: (i) higher resistivity values (~200 µΩcm) (ii) a low temperature resistivity minimum at certain temperature (T_{min}) and a broad maximum at higher temperatures (ii) the T_{min} shifts to lower temperatures and the broad maximum to higher temperatures with Co-content. Magnetic susceptibility ($\chi = dM/dH$) as a function of temperature, $\chi(T)$, (*Inset* of Fig. 1) shows a magnetic transition at lower temperatures (T_{peak}), which increases with Cobalt content (*Inset* of Fig. 3). The effect of magnetic field on the resistivity for a representative composition x = 54 is shown in Fig. 2. The magnetic field effects on $\rho(T)$ *i.e,* the T_{min} decreases on application of higher magnetic fields further supports (*Inset* (a) of Fig. 2) the above viewpoint. This results in progressively suppression of negative TCR on application of higher magnetic field. These observations suggest a correlation between magnetic and electrical properties, i.e, the T_{min} decreases with the increase in magnetization values. The $MR = [\rho(T, H = 90\,kOe) - \rho(TH = 0)]/\rho(T, H = 0)$ is plotted as a function of temperature for

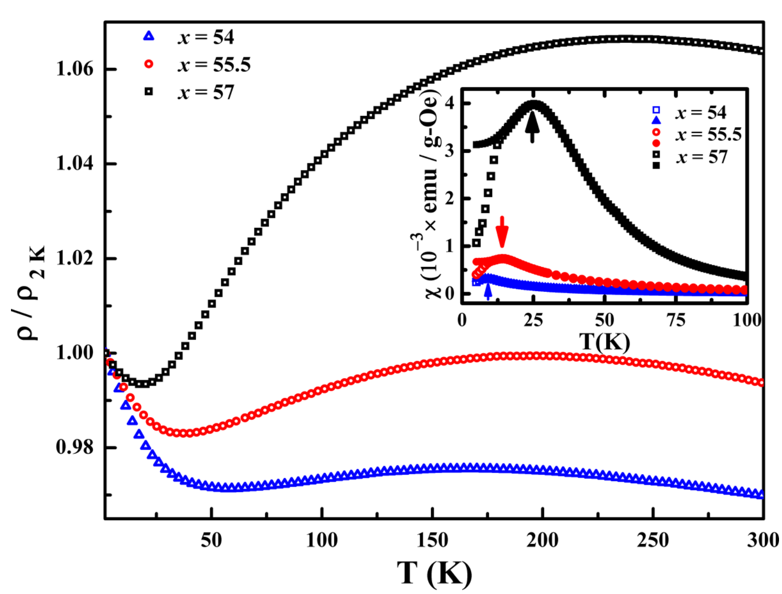

FIG. 1. Temperature variation of normalized resistivity for x = 54-57. *Inset*: Magnetic susceptibility (χ) as a function of temperature in zero field cooled and field cooled mode.

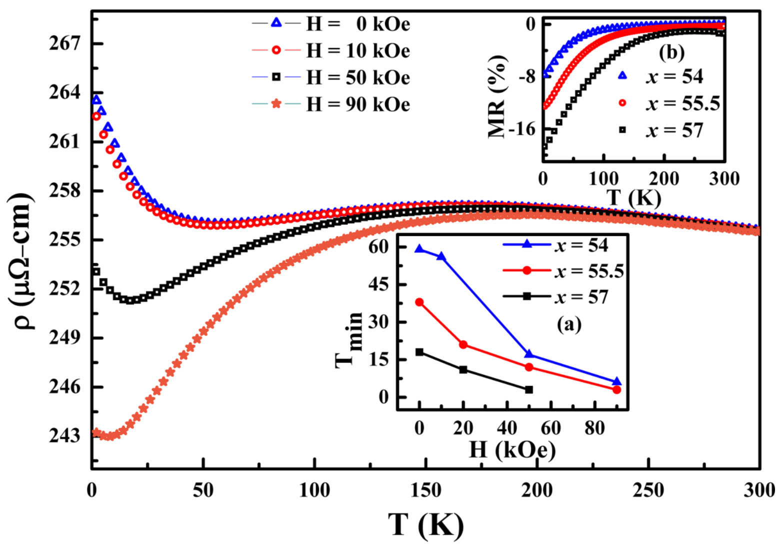

FIG. 2. Temperature variation of resistivity at different magnetic field for $x = 54$. *Inset* (a): $T_{\min}$ as a function of magnetic field for $x = 54$-57. Solid lines are guide lines to eye. *Inset* (b): MR as a function of temperature at an applied field of 90 kOe for $x = 54$-57.

all the compositions shown in *inset* (b) of Fig. 2. It is seen from the figure that significant MR is observed at very low temperature which rapidly vanishes with increasing temperature. The MR value is higher for $x = 57$ compared to those for $x = 54$ and 55.5 all over the temperature range. On the other hand higher resistivity values and negative TCR in crystalline Co-Ga suggest non-metallic behavior. Earlier experiments on high temperature resistivity results were interpreted on mainly two following aspects (i) temperature dependent Debye-Waller factor and the ω-phase transformation (ii) the band paramagnet homogeneous mixed-valency model and spin nature which consists of giant moments. Some of the earlier proposed models could not explain coherently the resistivity and magnetoresistance (MR) behavior observed in these compositions. It was suggested with the analogy to the Kondo problem that the electron-cluster spin interaction results in negative MR in Co_xGa_{100-x}. The low temperature resistivity anomalies in the present study resembles Kondo like behavior apparently, wherein the logarithmic divergent terms in the conduction-electron-localized spin exchange interaction give rise to a minimum in the resistivity at low temperatures when the exchange constant is negative. Further, the composition and field dependence of $T_{\min}$ supports this view point. However, when the resistivity data was fitted to logarithmic temperature dependence, the fits significantly deviated from linear behavior at very low temperature, though a linear tendency is observed at higher temperatures.

Earlier reports suggest that the deviations from stoichiometry results in structural defects in Co-Ga : vacancies on the Co sublattice and Co antistructure (AS) atoms on the Ga sublattice, these two types of defects being present even at the equiatomic composition. In weakly localized regime number of corrections, broadly classified as Weak Localization (WL) and Enhanced inter-Electron Interaction (EEI) to the transport coefficient of disordered conductors have been identified.[12] Such an approach can be implemented when the Fermi wavelength becomes comparable to the electronic mean free path. Based on the characteristic temperature-dependent corrections to conductivity, arising from localization and Coulomb interaction the resistivity can be expressed as[12]

$$\rho(T) = \frac{1}{\sigma_0 + aT^{p/2} + mT^{1/2}} + bT^2 \tag{1}$$

$aT^{p/2}$ is the localization term where the estimates for p are 3/2, 2, and 3, depending on whether Coulomb interactions in the dirty limit, clean limit, or electron-phonon scattering determine the inelastic scattering rate. The interaction term is proportional to $T^{1/2}$; but the sign of it's coefficient may change from positive to negative. In addition to these two corrections a term bT^2 is included

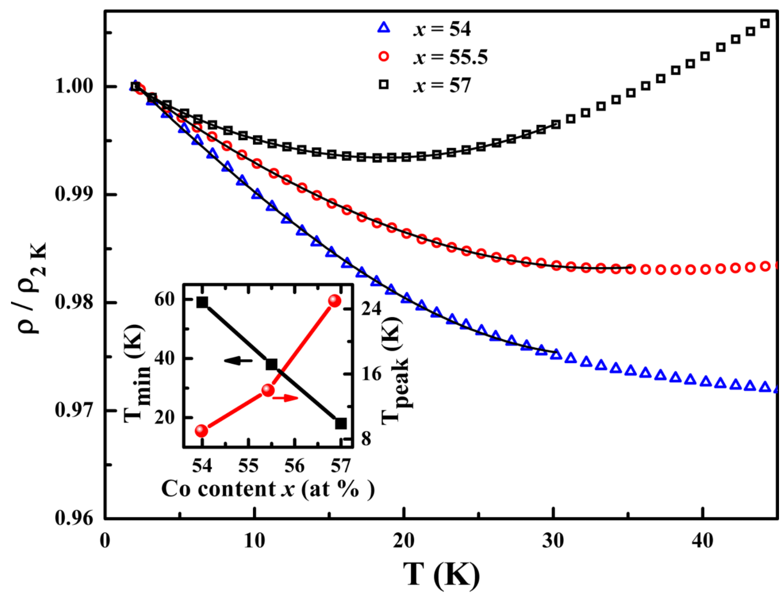

FIG. 3. Temperature variation of normalized resistivity at low temperature. Solid lines represent the fits to Eq. (1). *Inset*: T_{min} and T_{peak} as a function of Co content x.

to improve the quality of fits, which account for the relatively higher temperature scattering in the present system. If these mechanisms are assumed to be playing significant role in the transport phenomenon the Eq. (1) suggests that at the lowest temperatures, $\rho(T)$ is dominated by the interaction term because the index p is greater than 1. $\rho(T)$ plotted as a function of $T^{1/2}$ in the low temperature regime again shows a non-linearity raising a question whether Eq. (1) is inadequate to describe the transport properties of the present system. However, Eq. (1) describes the resistivity data of Co_xGa_{100-x} (x = 54-57) well up to a certain temperature range, with the exponent $p = 3$. For $x = 57$, the range of fit extends beyond T_{min}, whereas for $x = 54$ and 55.5 it is limited below T_{min} (Fig. (3)).

The application of magnetic field clearly demonstrates that residual resistivity is significantly reduced. However, with the application of strong magnetic field the range of fit is largely affected. In addition to this the same exponent value of p (= 3) cannot describe the transport properties of all the compositions (x = 54-57) satisfactorily.

We have attempted to interpret the low temperature magnetoresistance, MR(H), data with localized moment model. The localized moment concept provided good qualitative agreement with the observed concentration and temperature dependence of the negative magnetoresistance for some dilute alloys and degenerate semiconductor. For magnetic scattering of conduction electrons from localized magnetic moments Yosida, through 2$^{\text{nd}}$ order perturbation calculations, showed that in weak magnetic field MR is proportional to square of magnetization (M).[13] Therefore MR is plotted as a function of M^2 in the weak field regime as shown in *inset* of Fig. 4. It is observed that for $x = 54$ and 55.5 the behavior is somewhat similar and fairly linear behavior is observed which extends over much wider field compared to that for $x = 57$.

In order to account for the high field data we have attempted to implement the Khosla and Fischer model which considers higher order term (3$^{\text{rd}}$ order) to be equally contributing to MR as that of 2$^{\text{nd}}$ order term and experimentally observed in Cu-Fe and even in Cu-Mn alloys beyond the weak field regime.[14] A semiempirical expression of the form

$$\frac{\Delta\rho}{\rho_0} = -B_1 \ln(1 + B_2^2 H^2) \tag{2}$$

is used to fit the MR data. The fits to Eq. (2) resulted in deviations from the experimental data, more pronounced in $x = 57$ composition. Therefore, in addition to the localized moment model we

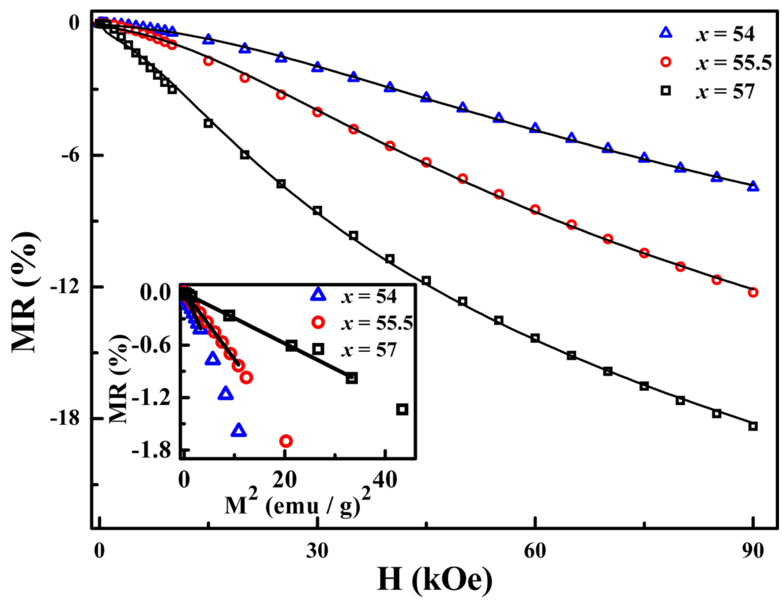

FIG. 4. MR at 5 K fitted to Eq. (2) in addition to the $H^{1/2}$ term. *Inset*: Low field MR data plotted as a function of M^2 at 5 K. The solid lines represent linear fits.

consider the WL and EEI effects with a view to interpret the MR data satisfactorily. In particular, magnetic impurities have two-fold effect on MR; a direct contribution due to the field dependence of the spin-flip scattering, and an indirect contribution from spin-flip dephasing of the electron wave function.[15] However, most experimental evidences manifested a poorer agreement with the theory in bulk disordered conductors compared to that in 2-D systems. The MR data at 5 K when plotted against $H^{1/2}$ and H^2 for high and low field, respectively shows fairly linear behavior, as expected from the asymptotic form in the expression for negative MR as suggested by Kawabata for 3-D WL regime.[16] Moreover, a combination of $H^{1/2}$ and $B_1 \ln(1 + B_2^2 H^2)$ (localized moment) is observed to fit the MR data well over a wide range of field (except very low field regime) as shown in Fig. 4.

The MR(H) decreases rapidly with the increase in temperature for $x = 57$ (Fig. 5) which is similar to that observed in granular materials. Further, as the Co concentration increases the MR

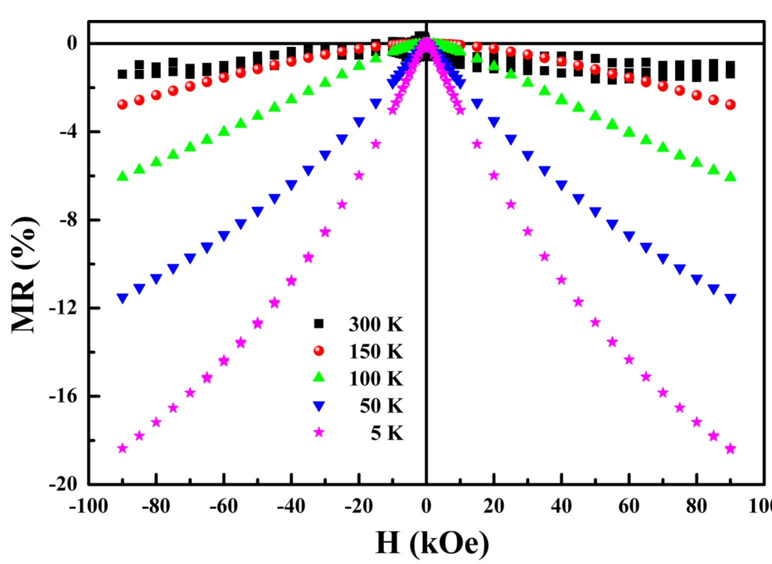

FIG. 5. Magnetoresistance as a function of applied fields at different temperatures for $x = 57$.

decreases and maximum MR is observed for $x \sim 57$ composition at 5 K. The equiatomic Co-Ga is nonmagnetic but with increase in Co-content the magnetic exchange interactions get enhanced with higher magnetization values and transition temperatures. This results in significant changes in transport behavior for $x \geq 58.5$, with the disappearance of low temperature minima and a significant reduction in MR, which also suggests the resistivity minimum is related to weak magnetic behavior.

Earlier reports suggested that the equi-atomic Co-Ga is a weak paramagnet whereas frustrated state has been identified above a certain Co concentration and $x \sim 57$ is reported as a critical concentration (x_c) beyond which ferromagnetic ordering develops. The low temperature MR value near x_c is comparable to that reported in GMR materials. Although the present system is not perfectly analogous to a granular systems, which primarily concerns an immiscible systems containing magnetic impurities in non-magnetic host (Co-Cu, Co-Ag, etc) the proposed model which attributes the magnetic behavior in the present systems only to clusters of AS Co atoms rather reinforces in describing the MR behavior in a similar manner.

IV. SUMMARY AND CONCLUSION

The low temperature electrical transport studies of the Co_xGa_{100-x} (x = 54-57) alloys have been discussed in detail. The resistivity anomalies observed in these off-stoichiometric compositions are due to magnetic impurity. The WL and EEI effects support the electrical transport properties of the system better than Kondo model. The large MR near the critical concentration could be due to development of ferromagnetic clusters. The field dependence of MR could be well represented by localized model with Quantum correction. Therefore the localized moment model that is used to explain the transport features of dilute magnetic semiconductors appears to be suitable for understanding the electrical transport properties of the present system.

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