On the Hysteresis Across First Order Paramagnetic-Ferromagnetic Transition in $Co(S_{1-x}Se_x)_2$, $0.07 \leq x \leq 0.14$

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Abstract. Hysteresis in resistivity across first order paramagnetic to ferromagnetic transition in polycrystalline$Co(S_{1-x}Se_x)_2$,$0.07 \leq x \leq 0.14$ have been studied. It shows that with increase in Se substitution transition temperature shifts to low temperature and hysteresis increases. As expected the hysteresis is predominantlydetermined by temperature. Though transition width depends sensitively on the disorder present in the sample, a monotonic increase in hysteresis is observed with Se substitution.

INTRODUCTION

CoS_2 undergoes second order magnetic phase transition from high temperature paramagnetic (PM) to low temperature ferromagnetic (FM) state with T_C= 124K[1, 2]. By substituting small amount of P, Se,As for S and Ni, Fe for Coin CoS_2, the transition temperature can be tuned over a wide temperature range[3, 4, 5, 6, 7]. Particularly Se substitution for S i.e. $Co(S_{1-x}Se_x)_2$ has been studied extensively [2, 8, 9] and these studies show decrease in $T_C$with x up to x = 0.1 and no transition for x >0.1. Around 1% Se substitution nature of transition becomes first order[9]. With Se substitution, unit cell volume increases, however its effect on $T_C$is reported to be similar to that of applying pressure such that 1% Se substitution equivalent to 0.51 GPa[9]. With pressure nature of transition changes to first order above around 0.4 GPa and finally no transition is observed for pressure ≥ 4.8 GPa[9, 10, 11]. It can be observed from the existing literature that hysteresis across first order transition increases with Se substitution [2, 8, 9]. Hysteresis has been one of the characteristic of first order transition which could be detrimental for many applications. Apart from hysteresis two phases separated by first order transition can co-exist at the transition. Due to finite correlation length there exist a distribution of $T_C$over sample volume on the length scale of correlation length. It leads to broadening of otherwise singular first order transition. Effect of pressure, magnetic field and temperature on hysteresis and transition width is studied by Kushwaha et al.[12] in Pd doped FeRh. It shows both hysteresis as well as transition width predominantly depends onthe temperature.

In this manuscript relative effects of substitution, magnetic field and temperature on hysteresis and transition width are reported. In case of substitution, there can be composition fluctuation across mean composition over sample volume. The degree of this fluctuation can vary from sample to sample. Present work shows that in spite of varying disorder or compositional fluctuation (evident from transition width)hysteresis increases with Se substitution.

EXPERIMENTAL TECHNIQUE

The polycrystalline samples with nominal composition$Co(S_{1-x}Se_x)_2$, $0.07 \leq x \leq 0.14$ are prepared by solid state reaction method [13, 14]. To check the phase purity, powder X ray diffraction measurements at room temperature are performed in Bruker D8 Advanced X-Ray diffractometer. Rietveld refinement using FullProfsoftware confirmed that all the samples crystallizes cubic pyrite structure and are single phase. Resistivity and magnetoresistance {MR= $[\rho(H) - \rho(0)]/\rho(0)$} measurements are performed using four probemethod in

longitudinal configuration in a home-made resistivity set up along with 8 T superconducting magnetic systems from M/s. Oxford Instruments,U.K.

RESULT AND DISCUSSIONS

Figure 1(a) shows temperature dependence of normalized resistivity (ρ/ρ_{300K}) for x = 0.07, 0.08 and 0.1 at zero field. With temperature cycling a hysteresis is observed which is taken as a signature of first order PM-FM transition. Generally, first order PM to FM transition is accompanied with decrease in resistivity due to decrease in spin disorder scattering.

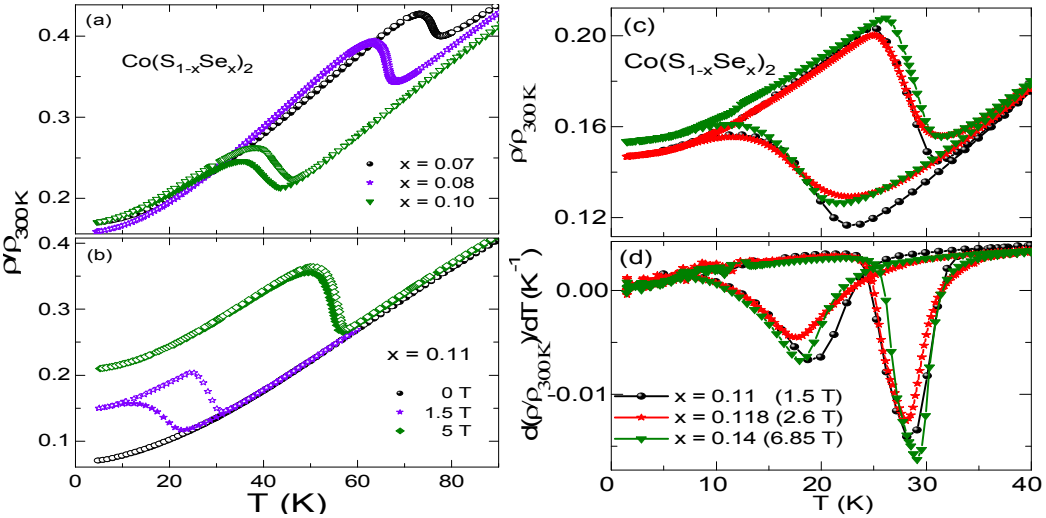

FIGURE 1(a)-(b)Temperature dependence of normalized resistivity (ρ/ρ_{300K}) for [a] x = 0.07, 0.08, 0.1 in the absence of applied magnetic fieldand [b] x = 0.11 in the presence of labeled magnetic field.(c)Shows temperature dependence of normalized resistivity (ρ/ρ_{300K}) and (d) its derivative for $Co(S_{1-x}Se_x)_2$ for the labeled x and H. (x, H) combinations are chosen so that transition temperature becomes nearly equal.

However in the case of CoS_2 system, it is accompanied withdistinct rise at T_C[1]. In the case parent CoS_2compound, FM state is reported to be highly spin polarized with 56-72% spin polarization[15, 16]. Therefore, higher resistivity in the FM state can be attributed to decrease in net density of states at the fermi level. Consistence with the literature resultstransition temperature (T_C) decreases with increase in x[2, 8, 9]. This figure 1(a) also highlights the increase in hysteresis with increase in x. The change in the hysteresis could be due to either change in x or different T_C.

Figure 1(b) shows data for x=0.11 in the presence of labeled magnetic field. Here, no transition is observed in the absence of magnetic field down to 5 K. In presence of magnetic field, field induce magnetic transition has been observed. The transition at 1.5 T during cooling from PM to FM state appears to be much broader compared to that observed during warming from FM to PM state. This could be due to lower transition temperature during cooling as compared with heating cycle. The value of resistivity at 5 K gradually increases with increase in magnetic field that could be due to increase in the FM phase fraction and it becomes nearly same above a certain field.

Moreover, figure 1(b) shows larger thermal hysteresis for 1.5 T curve in comparison to corresponding 5 Tesla data. Since composition is fixed it can be inferred that change in hysteresis is a consequence of applied magnetic field and temperature. In order to know the degree of disorder with Se substitution, residual resistivity ratio (RRR= ρ_{300K}/ρ_{5K}) is calculated. For CoS_2, it is found to be 63 and with Se substitution x=0.07, 0.08 and 0.1, this value is decreased to 5.86, 6.26 and 5.86.The RRR value for x=0.11, 0.118, 0.13 and 0.14 at 0 T are found to be 14.23, 13.32, 12.93 and 11.74 whereas in presence of 5 T for x=0.11, RRR value is found to be 4.76. The decrease of RRR value at 0 T implies degree of disorder increases with substitution.Now byusing opposing effect of x and H on $T_C$to compare hysteresis with nearly identical $T_C$but for different combination of (x, H). Result of such measurement is shown in figure 1(c), which shows quantitatively similar (ρ/ρ_{300K}) behavior for all the three compositions.

To compare the hysteresis and transition width, derivative of the curves in fig 1(c) are shown in figure 1(d). For this, the (T**-T*) is defined as the hysteresis (H_W) and (T*+T**)/2 as $T_C$where T* and T** are taken as peak value of the derivative of above ρ/ρ_{300K}-T data. Similarly transition width is taken as full width at half maximum of the derivative of above ρ/ρ_{300K}-T data. For a first order transition spatial distribution of T_C over sample volume due to composition fluctuations result in broadening of the transition. Therefore, width of the transition depends on quench disorder which can vary from sample to sample. Therefore the observed small change in the width of transition do not appear to follow systematic trend. Similarly the hysteresis is found to be 9.49 K, 10.2 K and 11.2 K for x=0.11 at 1.5 T, x=0.118 at 2.6 T and x=0.14 at 6.85 T respectively. Though small, it shows systematic increase in $H_W$with x. This trend in $H_W$should also reflect in field induced PM to FM transition. From figure 1, it appears that temperature plays dominant role in determining the hysteresis as compared to Se concentration.

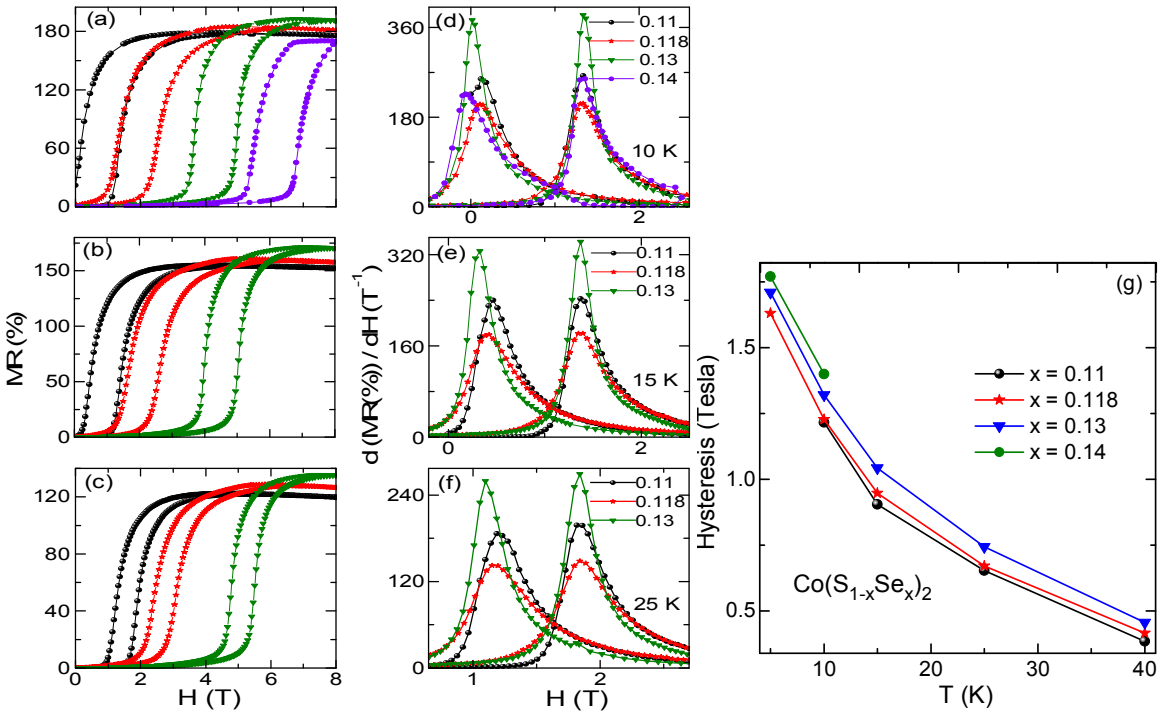

FIGURE 2 Isothermal MR (left panel) measured at [a] 10 K, [b] 15 K and [c] 25 K for $Co(S_{1-x}Se_x)_2$, x = 0.11, 0.118, 0.13, 0.14 and its derivative are shifted horizontally to match peak position of field increasing cycle with that of x=0.11 as shown in (d)-(f). [g] Shows temperature dependence of hysteresis estimated from above MR measurement.

To study the role of later (Se concentration) and to verify the trend in H_W, isothermal MR measurements arecarried out as shown in figure 2(a)-(c).It shows isothermal MR at various temperatures 10 K, 15 K and 25 K for various compositions x=0.11, 0.118, 0.13 and 0.14. Due to field induce magnetic transition it gives rise to giant positive magnetoresistance of more than 200%. At a given temperature, magnetic field required for PM to FM transition increases with Se concentration. It implies Se favors PM state. Here hysteresis is defined as the (H_{up}-H_{dn}) where $H_{up}$and $H_{dn}$are the peak value of the derivative of the isothermal magnetoresistance. For the sake of comparing the hysteresis of various compositions, these curves are shifted horizontally to match the H_{up}value of respective composition to that of x=0.11 and areshown in figure 2 (d)-(f). If temperature is the only factor which determines the hysteresis, then hysteresis is expected to same for all the compositions at a given temperature. However, figure 2 (d)-(f) shows that there is a systematicincrease in hysteresis with increase in x. The variation of hysteresis with temperature obtained from aboveMR data is plotted in figure 2 (g). It shows decrease in hysteresis with increase in temperature. It is expected as higher thermal energy facilitate transformation by crossing the energy barrier. Whereas correlation with composition (x) is not that straight forward. For example in the case of water to ice transition increased disorder provides nucleation site for the formation of ice and therefore hysteresis is reduced. Whereas, in the present system it shows opposite trend. There could be two possibilities for such behavior: (i) Unit cell volumechange across first order transition increases with increase in x thereby resulting in higher energy barrier

between two states at the T_C. The existing temperature dependent structural studies[2] in these system do indicate that for higher x the unit cell volume change at $T_C$increases. However, its x dependence for isothermal field induced transition is yet to be explored. (ii) Chaddah and Roy[17] addresses the issue ofobservable hysteresis with varying pressure/magnetic field. In their model they showed that the application of pressure (magnetic field) will lead to broadening of transition if the low temperature phase has lower density (magnetization) compared to the high temperature phase and vice versa. In the present system FM state has higher volume/magnetization and therefore hysteresis will increase with increase in substitution.

CONCLUSIONS

To conclude, hysteresis in resistivity across the first order paramagnetic to ferromagnetic transition in both temperature and magnetic field cycling for polycrystalline $Co(S_{1-x}Se_x)_2$, $0.07 \leq x \leq 0.14$ have been studied. With increase in substitution at zero field hysteresis in resistivity increases that could be due to both substitutional disorder and temperature effect. There is though small but a systematic increase in the hysteresis with decrease in temperature has been observed in isothermal MR. One parameter could be unit cell volume change at the transition, which increases the potential barrier height across the paramagnetic (PM) to ferromagnetic (FM) transition. High field X-ray diffraction studies will be useful to address this issue.

ACKNOWLEDGMENTS

I thank R. Rawat for discussion and fruitful suggestion.

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