Magnetic ordering and frustration in $Ge_{1-x-y}(Sn_xMn_y)Te$ multiferroic crystals

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Abstract

- Incorporation of magnetic ions in group IV-VI ferroelectric-semiconductor lattice offers intriguing materials properties and entanglement between magnetic and spin-orbit orders.
- $Ge_{1-x}TM_xTe$ is proposed to integrate its intrinsic ferroelectric polarization (broken inversion symmetry) and incorporated magnetic order which yields to Rashba spin splitting.
- We present $Ge_{1-x-y}(Sn_xMn_y)Te$ multiferroics to study its magnetic ordering, ferroelectric polarization and its domain walls dynamics at various temperatures.
- The studied samples exhibit spin-glass like ordering for $y = 0.047$. Both frequency dependent susceptibility $Re\chi(T)$ and coercivity $H_C(T)$ illustrate cusps near T_F.
- Anomalous Hall effect was observed which demonstrate dependency on magnetic field similar to magnetization curves below 0.1 T, figure is shown for $y = 0.027, 0.061$

Introduction and motivation

Why we chose IV-VI semiconductors:

- Higher solubility of transition metal (TM) ions [1].
- Spontaneous ferroelectricity and narrow band gap, crucial for Rashba splitting of spin bands.
- High T_c of 200 K for Mn = 0.5 [2].
- Ferroelectric nature of GeTe offers possibility of entanglement between magnetic and spin-orbit interaction [3].
- The temperature dependent ferroelectric transition offers studies of domain walls and its charge dynamics.

Motivation:

- **We are exploring GeTe based diluted magnetic semiconductors to study correlation between their ferroelectric and magnetic ordering.**
- **Various compositions of $Ge_{1-x}TM_xTe$ are being investigated to probe the ferroelectric transition temperature and its temperature dependent features.**
- **In this study, our proposed work is also focused on the low temperature ferroelectric domain walls dynamics for atomic circuitry applications.**

[1] L. Kilanski et al. *Phys. Rev. B* **95**, 035206 (2017).
[2] Y. Fukuma et al. *Appl. Phys. Lett.* **93**, 252502 (2008).
[3] J. Krempasky´ et al. Nature Communications 7 (1), 1-7 (2016).

Rashba splitting in IV-VI semiconductors

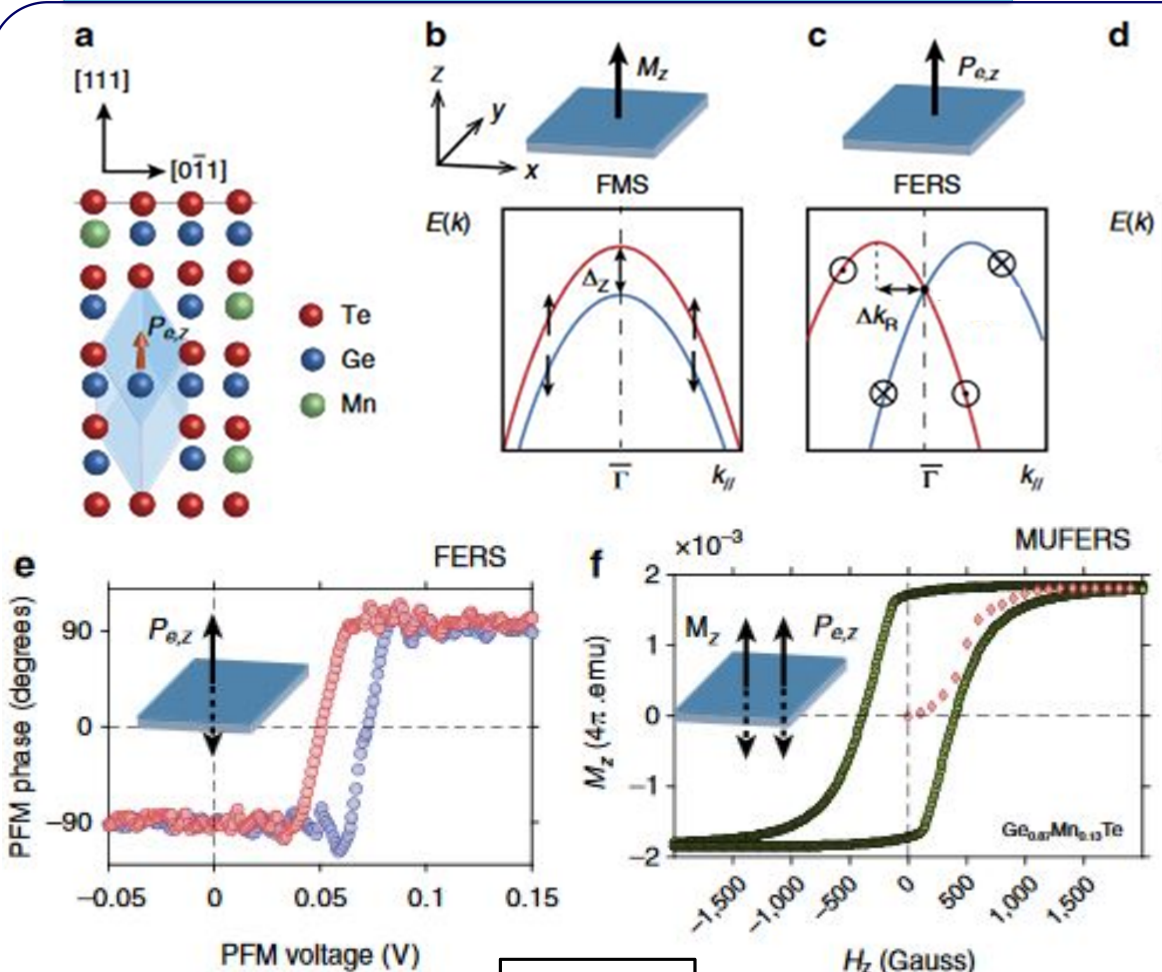

Figure 1

(a) Rhombohedrally distorted unit cell of multiferroic $Ge_{1-x}Mn_xTe$ (b) Schematic Rashba-gas band maps of a ferromagnetic semiconductor compared with ferroelectric Rashba semiconductor (FERS) (c) multiferroic Rashba semiconductor (MUFERS) with (d) Their dependence on the orientations of the FM (M) and FE (P_e) order (e) Typical out-of-plane FE phase hysteresis measured by piezo-force-micros-copy for (FE) GeTe and $Ge_{1-x}Mn_xTe$ (f) Out-of-plane FM hysteresis curve of multiferroic $Ge_{0.87}Mn_{0.13}Te$ measured by SQUID [4].

[4] J. Krempasky´ et al. Nature Communications 7 (1), 1-7 (2016).

Lattice structure

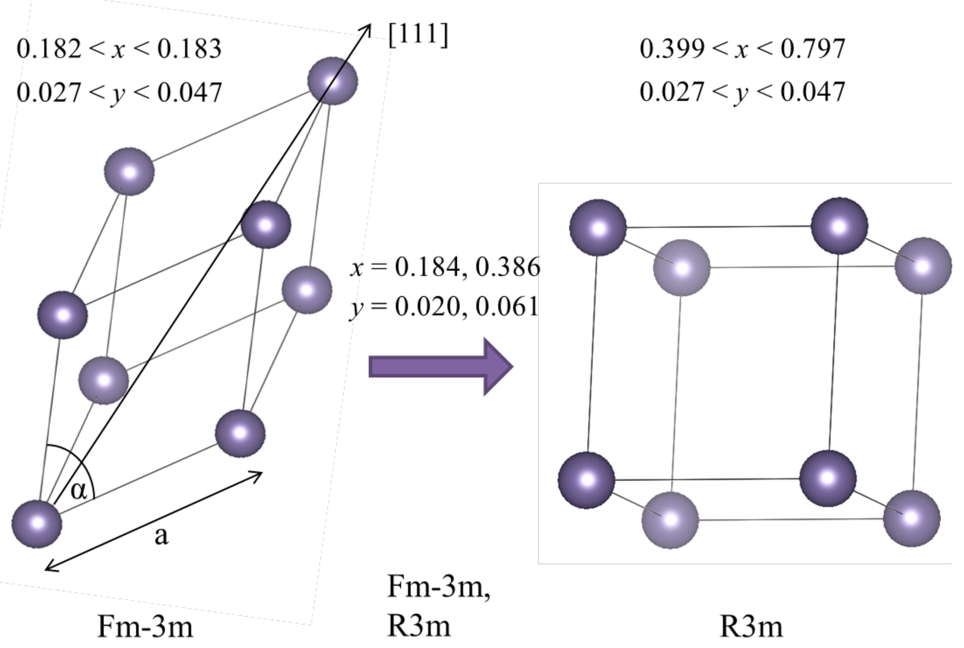

Figure 2

- The parent alloy α-GeTe has a rhombohedral (Fm-3m) structure below 720 K.
- $Ge_{1-x-y}Sn_xMn_yTe$ crystals preserve Fm-3m symmetry for lower dopant contents manifesting multiferroic features at room temperature.
- The high solubilities of IV-VI alloys for transition metals originate from the Te-5p anti-bonding states which are favorable to acceptor doping [5].

[5] T. Fukushima et al. *J. Phys. Condens. Matter* **27**, 015501 (2015) .

Magnetic properties

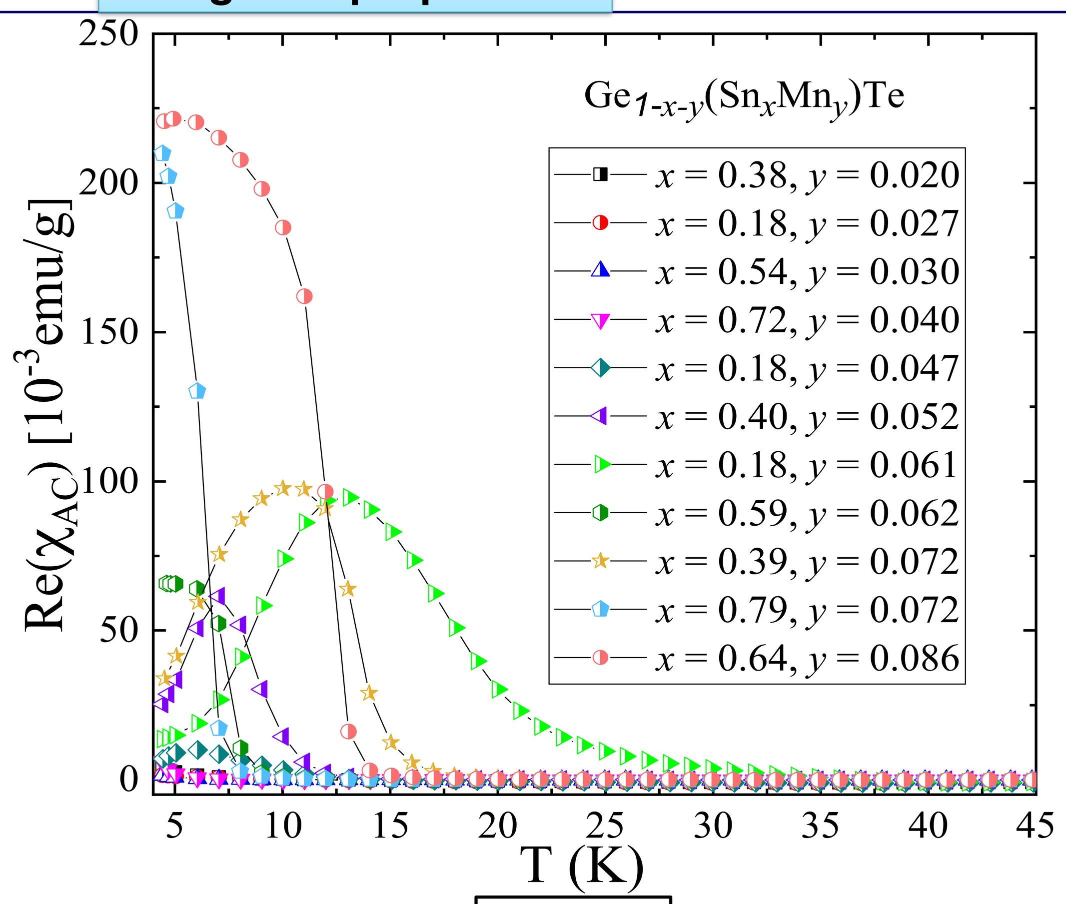

Figure 3

- Magnetic susceptibility graphs show different magnetic orderings between 4.5 and 25 K.
- The symmetric cusps indicate crystals which may have spin-glass or superparamagnetic ordering.
- Crystals such as $x = 0.72$, $y = 0.04$ exhibit a paramagnetic-like state.

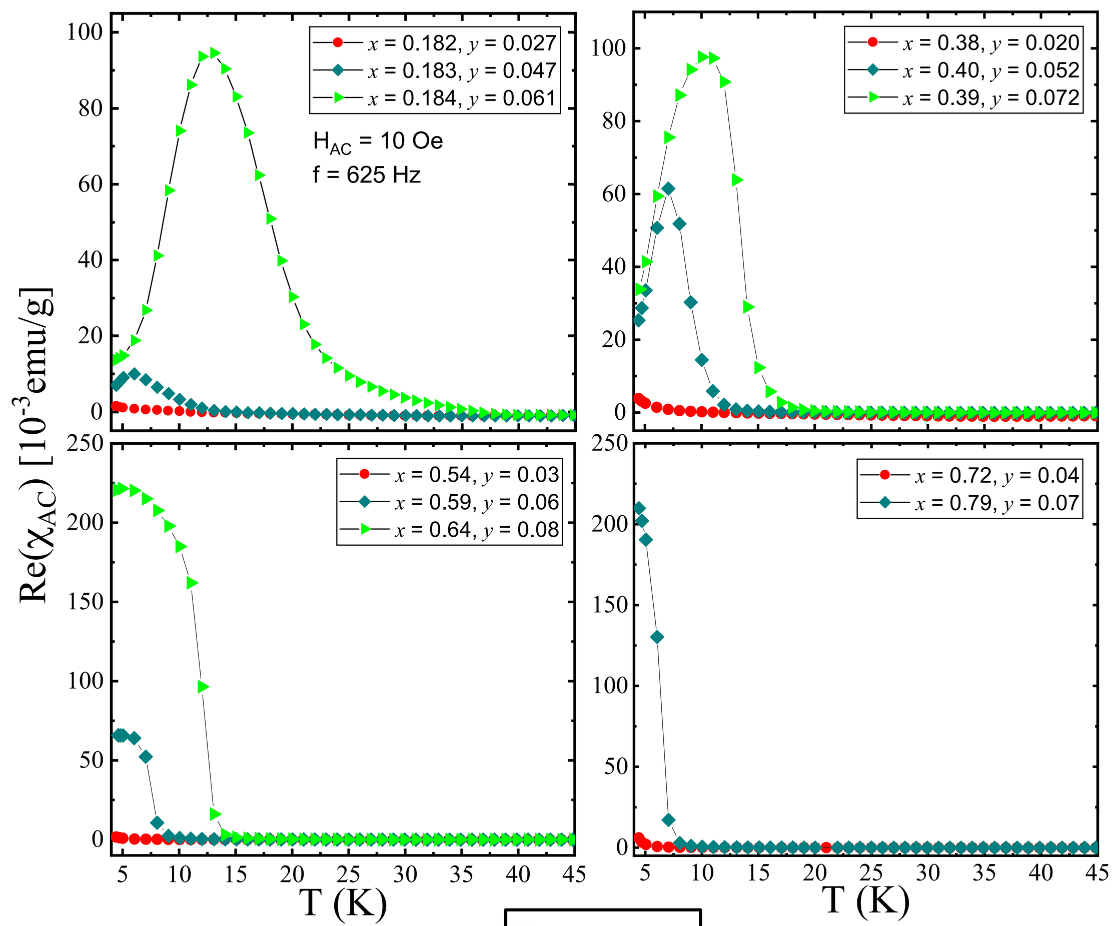

Figure 4

- We observad a spin-glass or superparamagnetic ordering for $0.2 < x < 0.4$ and $0.02 < y < 0.07$.
- However, the crystals display a ferromagnetic ordering for $0.5 < x < 0.8$ and $0.2 < y < 0.085$

Magnetic properties

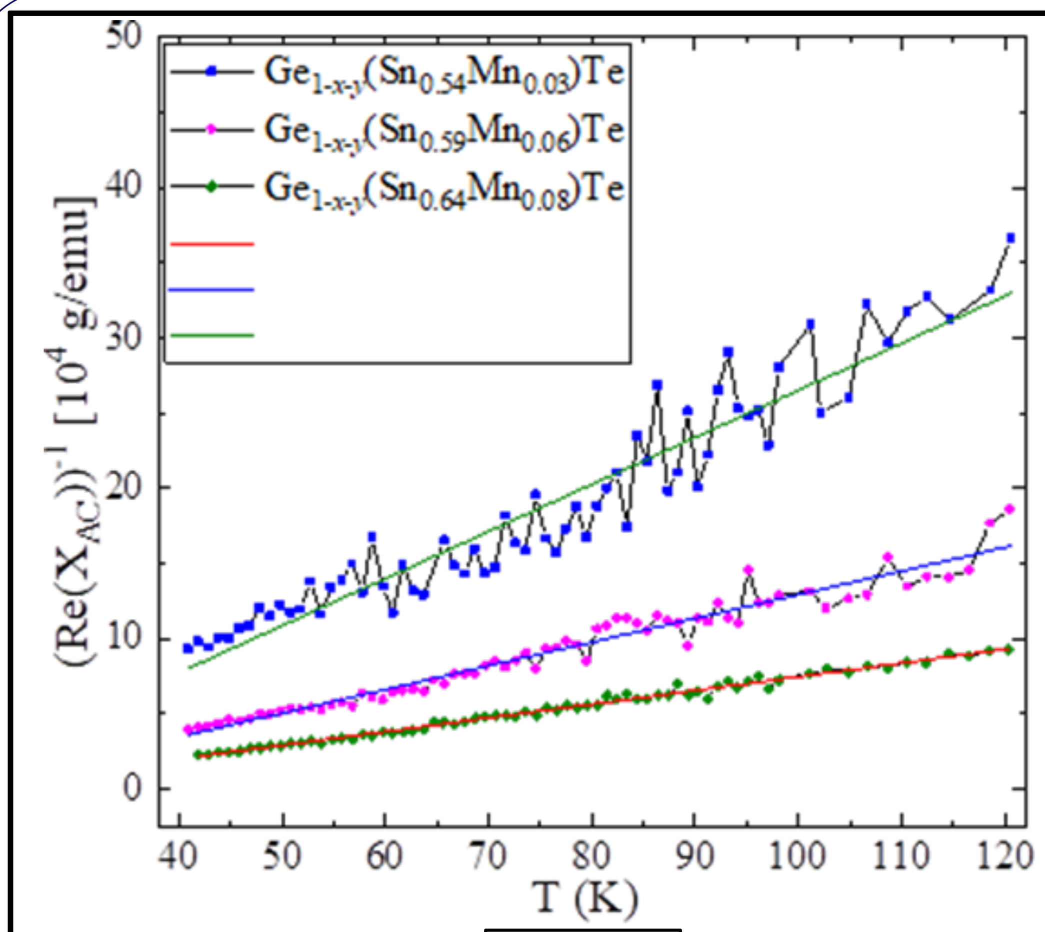

Figure 5

- Modified Curie-Weiss law was used to fit the paramagnetic region of the susceptibility,

$$\chi_i(T) = \frac{C}{T-\Theta} + \chi_{dia}$$

- Diamagnetic component, $\chi_{dia} \approx -3 \times 10^{-7}$ emu/g.

- Magnetization curves exhibit a square-like hysteresis for $y = 0.061$ indicating a ferromagnetic ordering.
- For $y = 0.047$, a cusp appears in coercivity near T_F similar to the frequency dependent susceptibility cusp.
- The cusp in $H_C(T)$ reflects variations in the magnetic disorder which might occur in the current spin-glass like crystal around its freezing temperature

Spin-glass like features

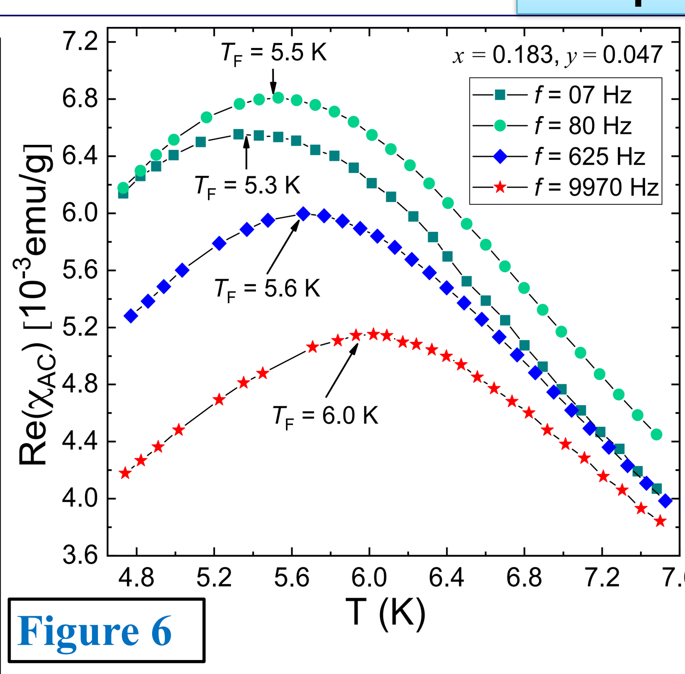

Figure 6

- Frequency dependent susceptibility for $x = 0.23$, $y = 0.05$.
- Freezing temp shifts from 5.5 K (7 Hz) to 6 K (9970 Hz).
- Calculated scaling parameter, **R** = 0.017 indicates spin-glass like ordering.

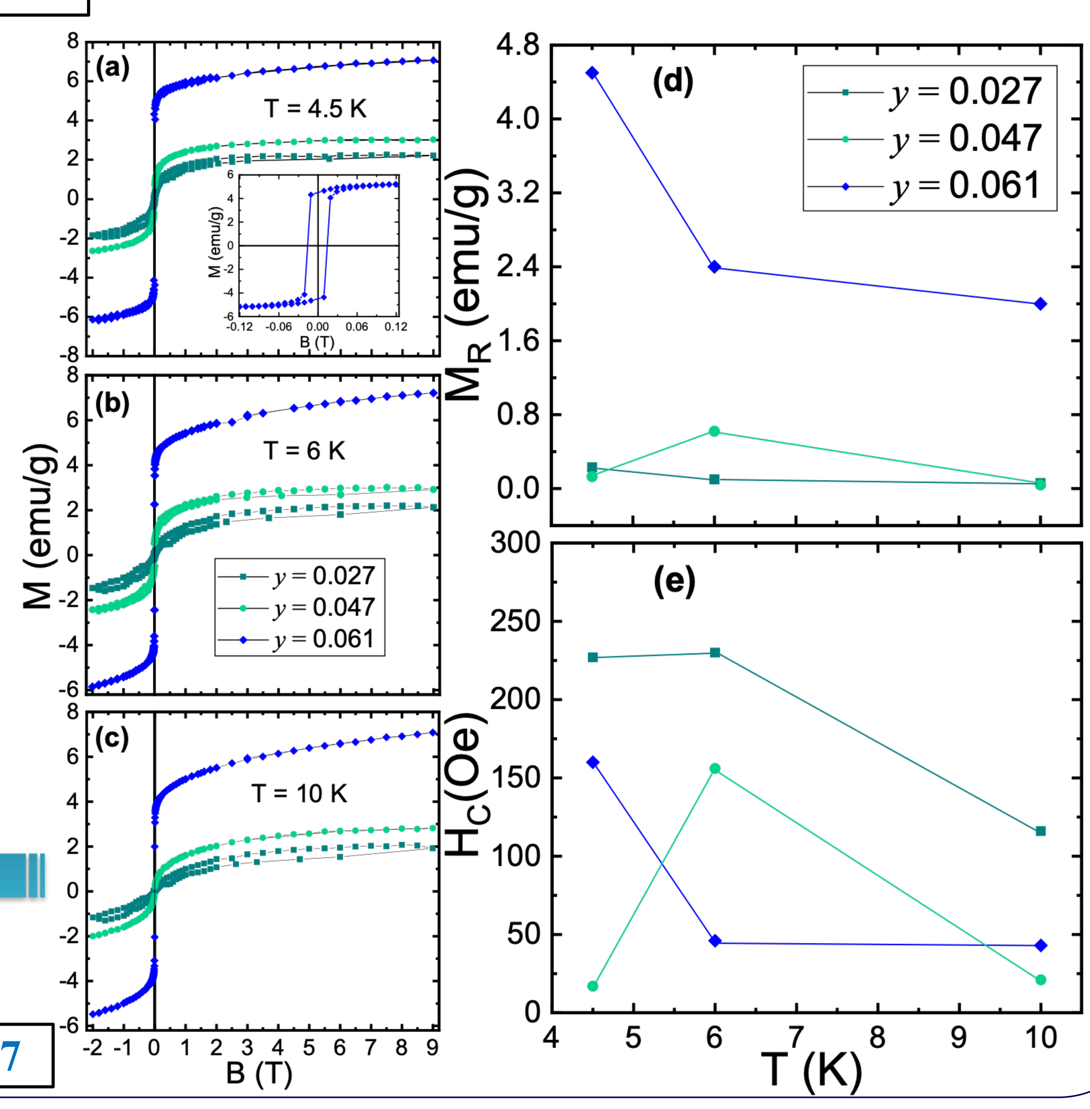

Figure 7

Anomalous Hall effect

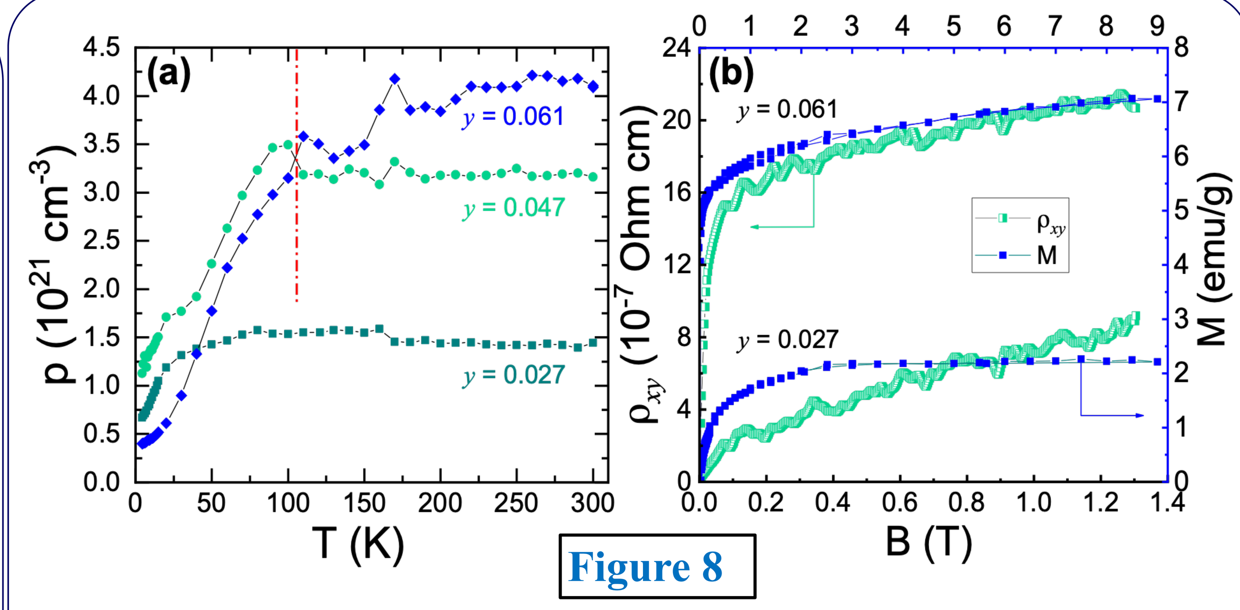

Figure 8

- The carrier concentration $p(T)$ plots behave differently below and above ≈ 100 K for y = 0.047, 0.061, Fig 8(a).
- The $p(T)$ plots indicate a degenerate semiconductor behavior above ≈ 100 K.
- Large anomalous Hall effect (AHE) is observed, see Fig 8(b).
- The anomalous Hall effect is dominant where the magnetization has linear dependency below a similar B-value.

Conclusions

- $Ge_{1-x-y}Sn_xMn_yTe$ semiconductor multiferroic samples were prepared using modified Bridgman growth method.
- The crystalline structure switches from rhombohedral to cubic symmetry for higher doping contents of Sn and Mn.
- The magnetic susceptibility measurements show both ferromagnetic and spin-glass like ordering, followed by similar cusp in $H_C(T)$ graph.
- The carrier concentration show a degenerate semiconductor behavior above the AHE region i.e. for T >100 K.
- Off-diagonal $\rho_{xy}(B)$ curves display large anomalous Hall effect.