# Magnetic ordering in $Ge_{1-x-y}(Sn_xMn_y)Te$ multiferroics

Lukasz Kilanski [1,*] Abdul Khaliq [1], Monika Arciszewska [1], Andrzei Avdonin [1], Beata Brodowska [1], Abbas Khan [1], Witold Dobrowolski [1], Vasya E. Slynko [2], Evgen I. Slynko [2],

[1] Institute of Physics, Polish Academy of Sciences, al. Lotnikow 32/46, 02-668 Warsaw, Poland.

[2] Institute of Materials Science Problems, Ukrainian Academy of Sciences, Chernovtsy, Ukraine.

Correspondence to: abdul.khaliq@ifpan.edu.pl

## Abstract

❖ Incorporation of magnetic ions in group IV-VI ferroelectric-semiconductor lattice offers intriguing materials properties and entanglement between magnetic and spin-orbit orders.
❖ GeTe is proposed to integrate both its intrinsic ferroelectric polarization (broken inversion symmetry) and incorporated magnetic order which yields to Rashba spin splitting.
❖ We have synthesized $Ge_{1-x-y}(Sn_xMn_y)Te$ multiferroics to study its magnetic ordering, ferroelectric polarization and its domain walls dynamics at various temperatures.
❖ Our measured samples show that $Ge_{1-x-y}(Sn_xMn_y)Te$ has cubic (Fm-3m) and rhombohedral (F3m) symmetries at room temperature for different Sn and Mn contents.
❖ The magnetic susceptibility measurements demonstrate the existence of both ferromagnetic and frustrated magnetic ordering at low temperature.

## Introduction and motivation

**Why we chose IV-VI semiconductors:**

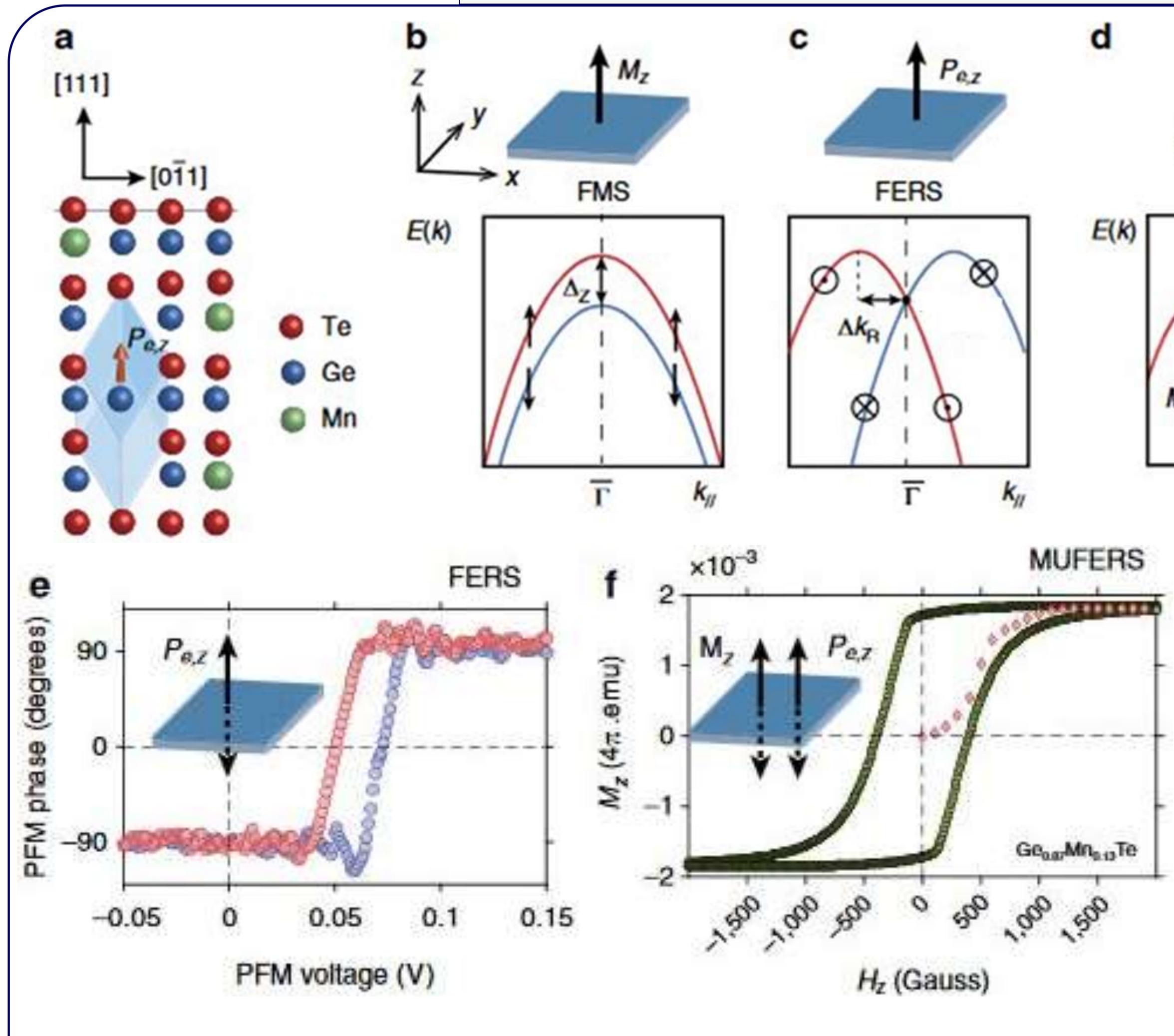
❖ Higher solubility of transition metal (TM) ions [1].
❖ High carrier concentration compared to II-VI compounds.
❖ $T_c = 200$ K for $x = 0.5$ [3].
❖ Ferroelectric ordering in $Ge_{1-x}TM_xTe$ [1-3].
❖ Strong RKKY interaction.
❖ Ferroelectric nature of GeTe offers possibility of entanglement between magnetic and spin-orbit orders, and Rashba spin splitting.

[1] L. Kilanski et al. *Phys. Rev. B* **95**, 035206 (2017).
[2] M. Hassan et al. *J. Cryst. Growth* **323**, 363 (2011).
[3] Y. Fukuma et al. *Appl. Phys. Lett.* **93**, 252502 (2008).

**Motivation**

- **We are exploring GeTe based diluted magnetic semiconductors to study their ferroelectric and magnetic ordering.**
- **Various compositions of $Ge_{1-x}TM_xTe$ are being investigated to construct the phase diagram based on ferroelectric transition temperature.**
- **In this study, our planned work is also focused on the low temperature ferroelectric measurements and dynamics of domain walls.**

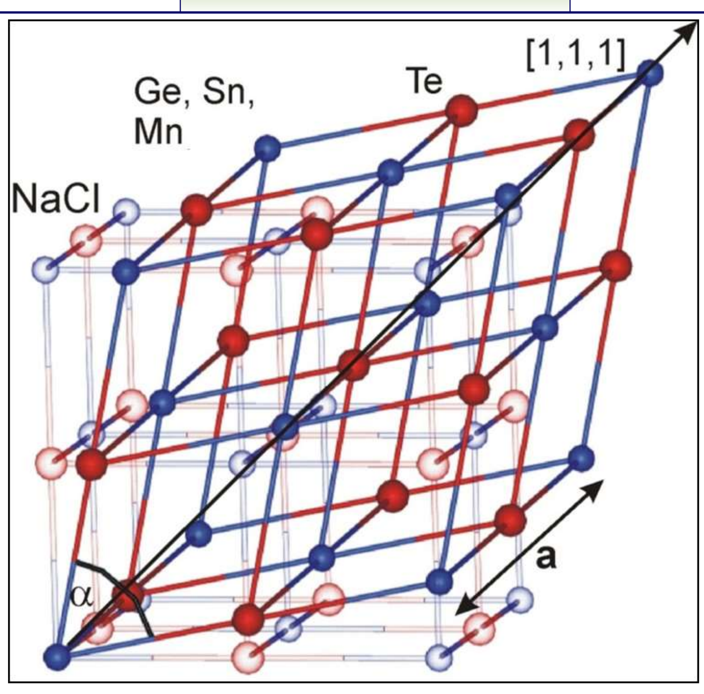
## Rashba splitting in IV-VI semiconductors




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

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) in d, with 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 preserved in $Ge_{1-x}Mn_xTe$ (f) Out-of-plane FM hysteresis curve of multiferroic $Ge_{0.87}Mn_{0.13}Te$ measured by SQUID [4].

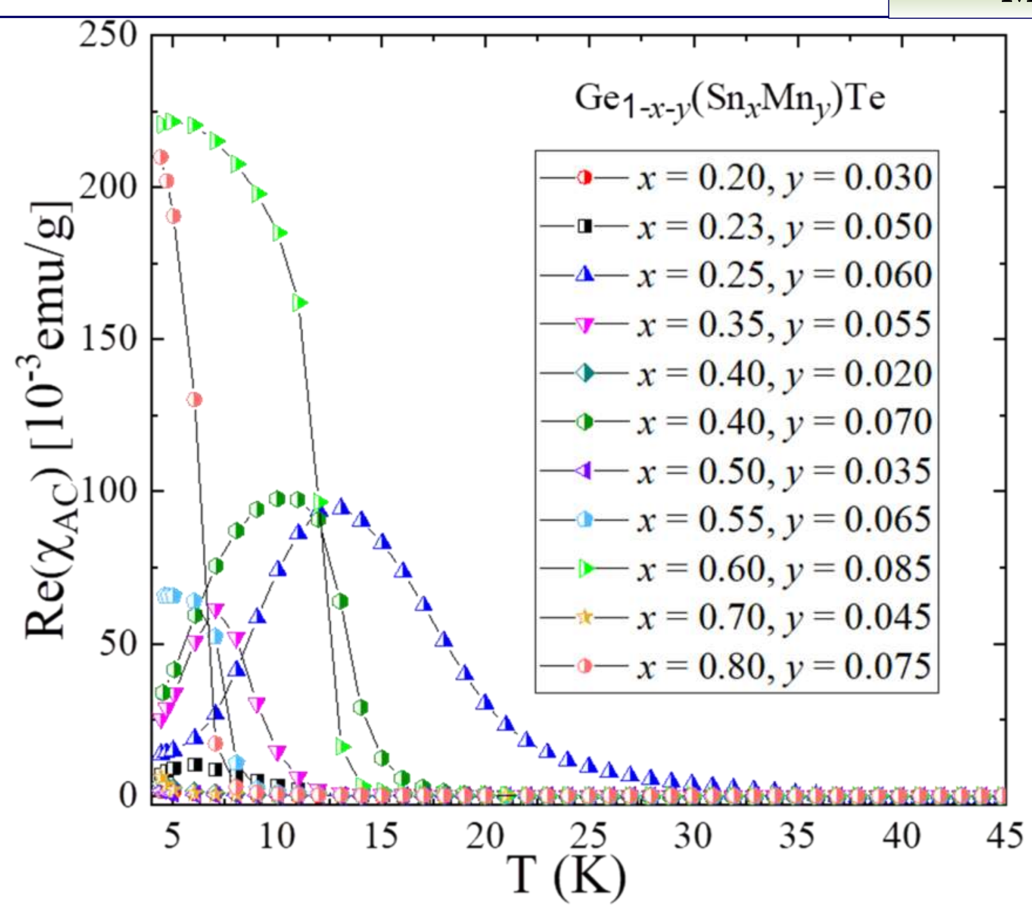
## Lattice structure



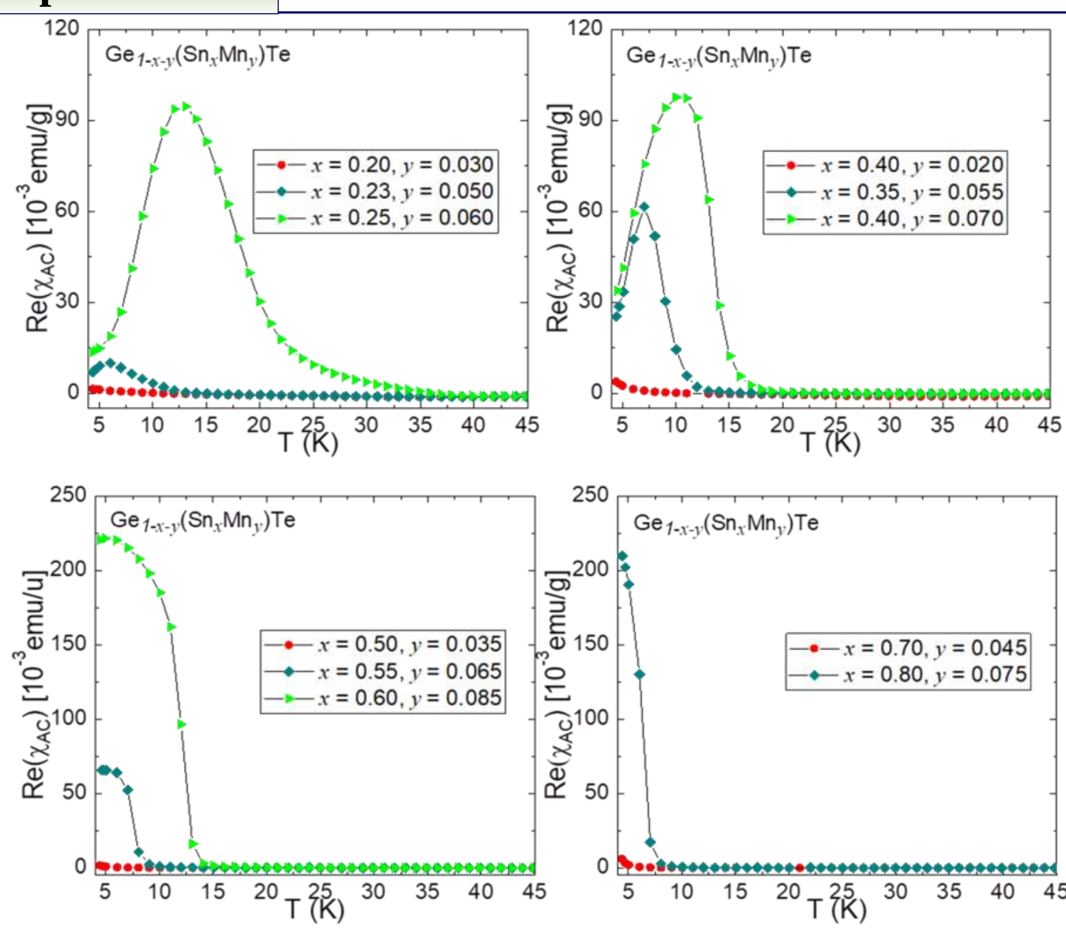

❖ Semiconductor-GeTe has NaCl like cubic structure.
❖ $Ge_{1-x-y}Sn_xMn_yTe$ reveals distortion in (111) direction indication a rhombohedral symmetry.
❖ IV-VI structures have high solubility for transition metal (TM) ions.
❖ The high solubilities originate from the Te-5p anti-bonding states which are favorable to acceptor doping [5].

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

## Magnetic properties

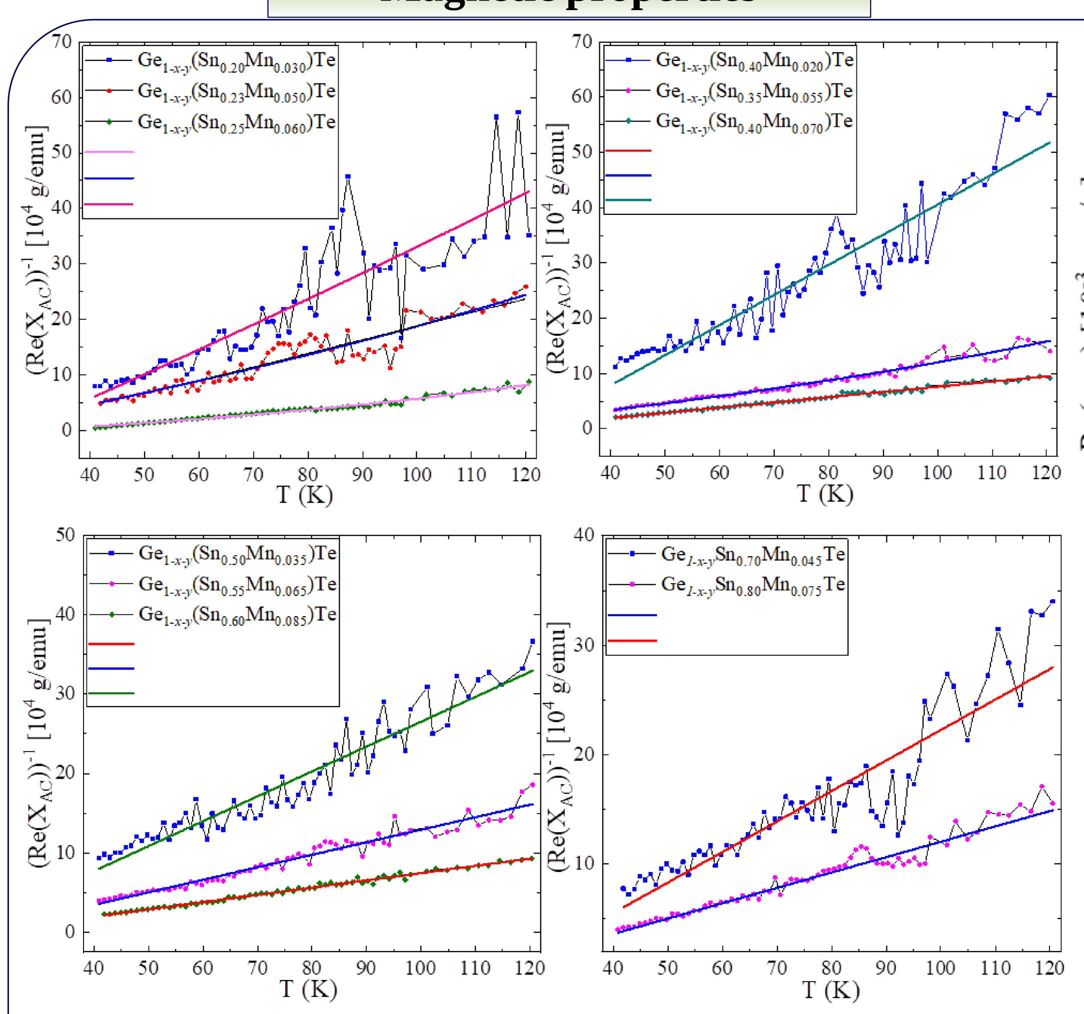



❖ Magnetic susceptibility measured from 4.5-300 K.
❖ The susceptibility graph shows both ferromagnetic and frustrated magnetic states.




❖ The susceptibility graphs indicate frustrated magnetic order for $0.2 < x < 0.4$ and $0.02 < y < 0.07$
❖ Whereas for $0.5 < x < 0.8$ and $0.035 < y < 0.085$ range, it exhibits a ferromagnetic ordering

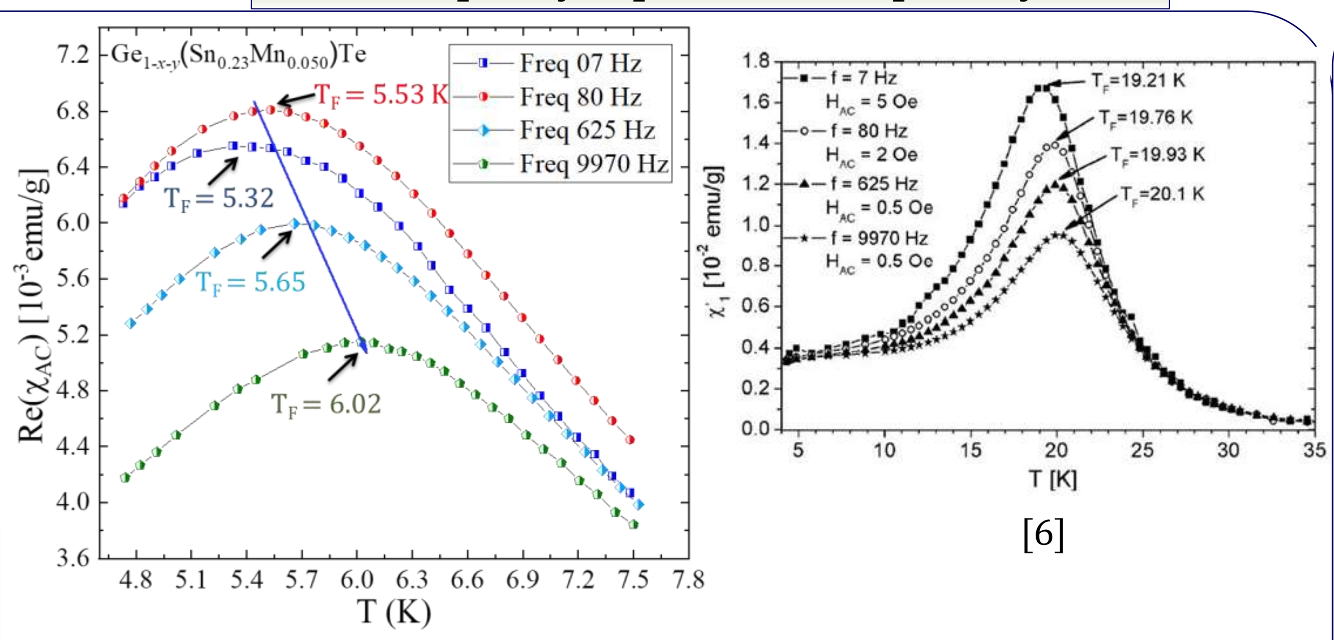
## Magnetic properties




❖ Modified Curie-Weiss law, was used to fit the paramagnetic region of susceptibility.
❖ $\chi_{dia} \approx -3 \times 10^{-7}$ emu/g was used for the diamagnetic GeTe.
❖ Modified Curie-Weiss law, $\chi_1(T) = \frac{C}{T-\Theta} + \chi_{dia}$

[6] L. Kilanski et al. *J. Appl. Phys.* **105**, 103901 (2009).

## Frequency dependent susceptibility




[6]

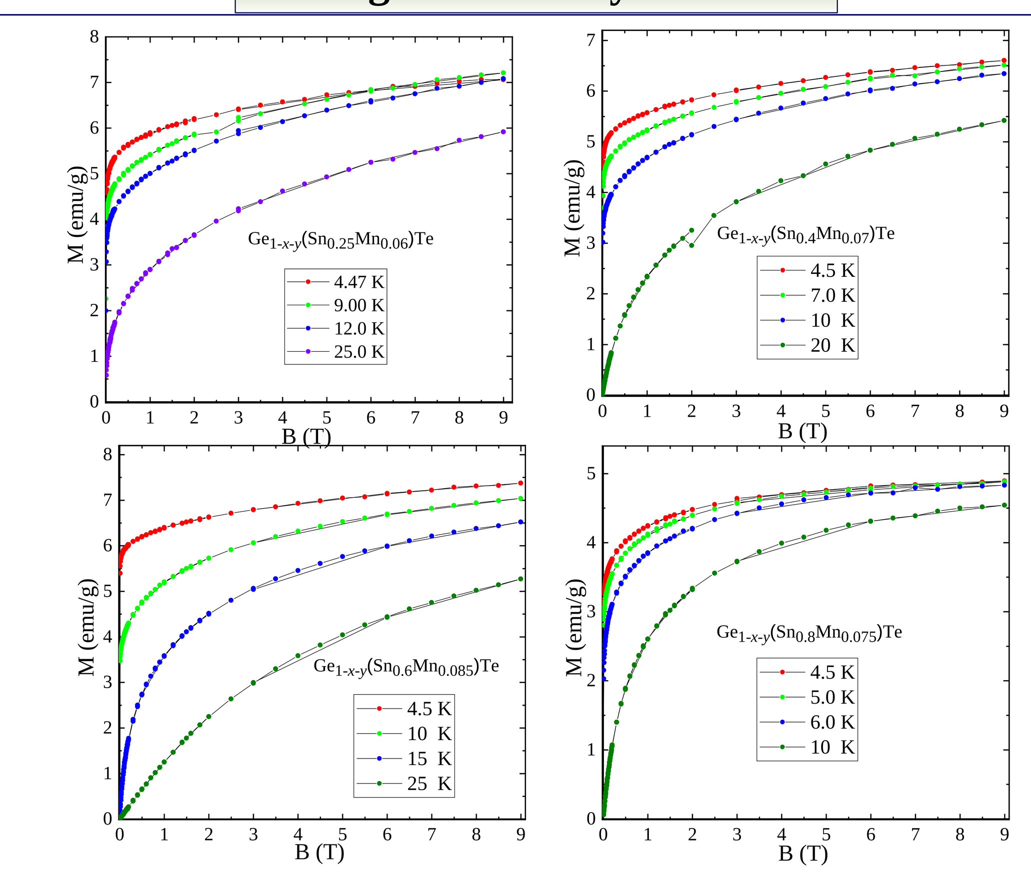
❖ Frequency dependent susceptibility was measured for x = 0.23, y = 0.05.
❖ Freezing temp shifts with change in frequency.
❖ Calculated Mydosh parameter, R = 0.017, indicating spin-glass like ordering

Using modified Curie-Weiss law fitting, the Curie-Weiss temperature Θ and Curie-Weiss constant C values were calculated, given in the table 1.

| Sn ($x$) | Mn ($y$) | Θ [K] | C ($10^{-4}$) [emu.K/g] |
|---|---|---|---|
| 0.20 | 0.030 | 26.6 ± 1.2 | 2.35 ± 0.8 |
| 0.23 | 0.050 | 25.6 ± 1.1 | 3.65 ± 0.8 |
| 0.25 | 0.060 | 40.6 ± 1.4 | 10.1 ± 1.5 |
| 0.35 | 0.055 | 23.6 ± 1.7 | 6.8 ± 1.3 |
| 0.40 | 0.020 | 17.9 ± 1.8 | 2.54 ± 0.7 |
| 0.40 | 0.070 | 19.7 ± 1.9 | 10.5 ± 1.1 |
| 0.50 | 0.035 | 25.9 ± 1.9 | 2.85 ± 0.2 |
| 0.55 | 0.065 | 18.4 ± 1.4 | 6.45 ± 0.6 |
| 0.60 | 0.085 | 17.8 ± 1.2 | 11.1 ± 1.3 |
| 0.70 | 0.045 | 30.3 ± 1.5 | 3.2 ± 0.2 |
| 0.80 | 0.075 | 15.3 ± 1.5 | 7.7 ± 1.2 |

Table. 1

## Magnetization hysteresis




❖ Magnetization measurements at different temperatures show a very narrow hysteresis.
❖ Showing the transition to a paramagnetic region.

## Conclusion

❖ $Ge_{1-x-y}Sn_xMn_yTe$ semiconductor multiferroic samples were prepared using modified Bridgman growth method.
❖ The crystalline structure indicate both cubic and rhombohedral symmetries, signature of spontaneous ferroelectric polarization.
❖ The magnetic susceptibility measurements show both ferromagnetic and frustrated magnetic ordering.
❖ Frequency dependent susceptibility shows a shift in freezing temperature ($T_F$) at various frequencies.
❖ The calculated Mydosh parameters for frequency dependent plots demonstrate a spin-glass like magnetic ordering.