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Simultaneously enhanced mechanical and thermoelectric performance of Ag_9GaSe_6 argyrodites via tailoring chemical compositions

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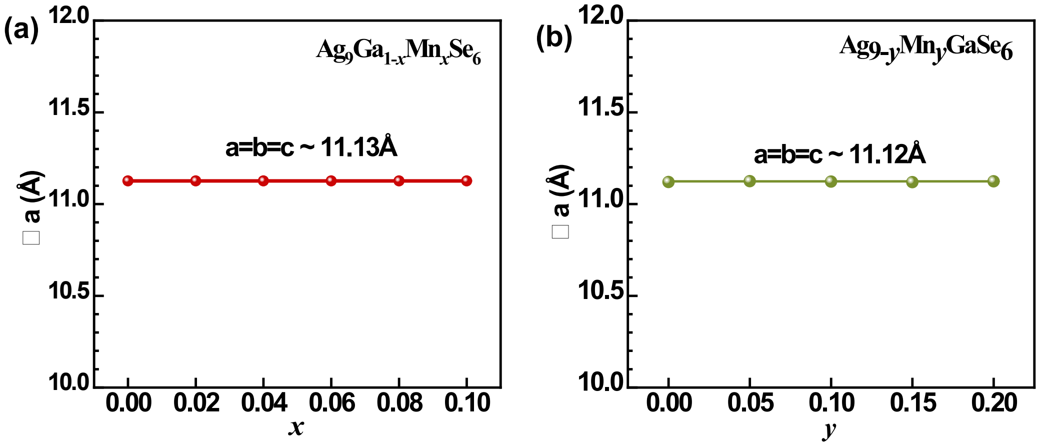

Figure S1. The estimated lattice constants for (a) $Ag_9Ga_{1-x}Mn_xSe_6$ and (b) $Ag_{9-y}Mn_yGaSe_6$ samples.

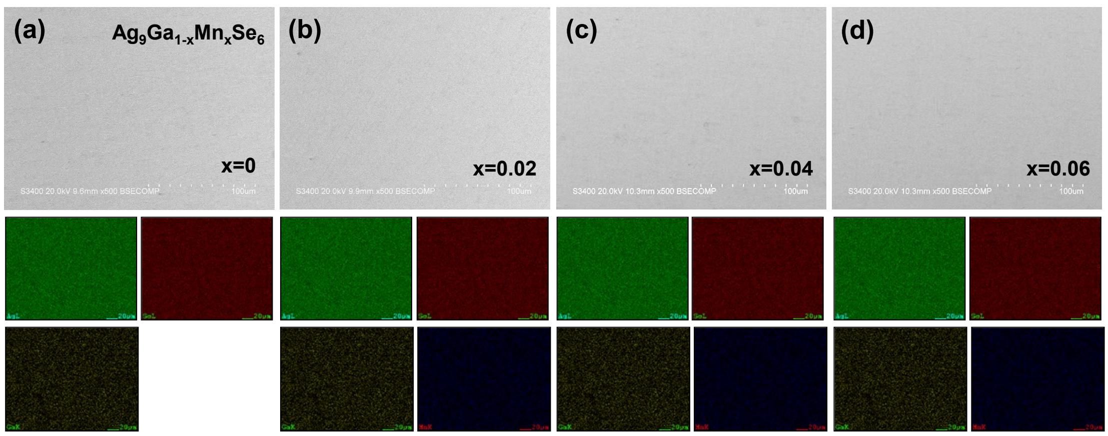

Figure S2. SEM image and corresponding EDS mapping for (a-d) $Ag_9Ga_{1-x}Mn_xSe_6$ samples with (a) x=0, (b) x=0.02, (c) x=0.04, (d) x=0.06, respectively.

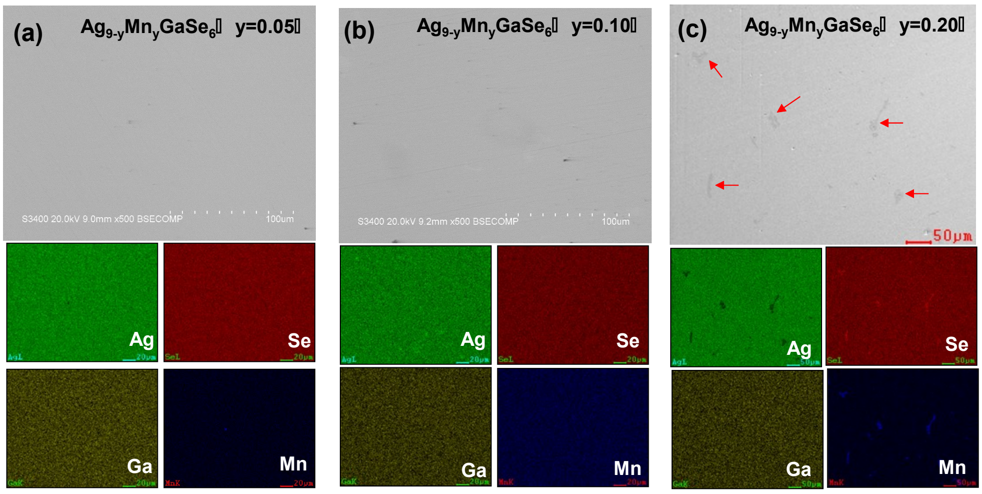

Figure S3. SEM image and corresponding EDS mapping for $Ag_{9-y}Mn_yGaSe_6$ samples with (a) y=0.05, (b) y=0.10, (c) y=0.20, respectively.

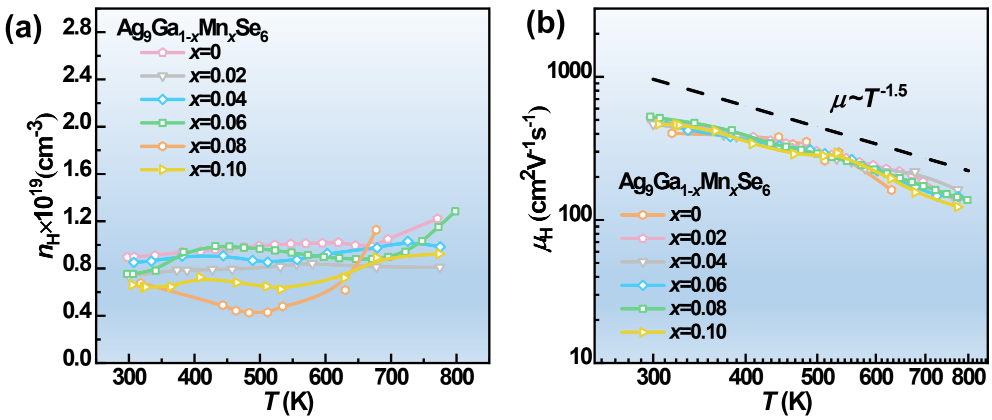

Figure S4. Temperature dependent (a) Hall carrier concentration and (b) Hall mobility for $Ag_9Ga_{1-x}Mn_xSe_6$ (0≤x≤0.10) samples.

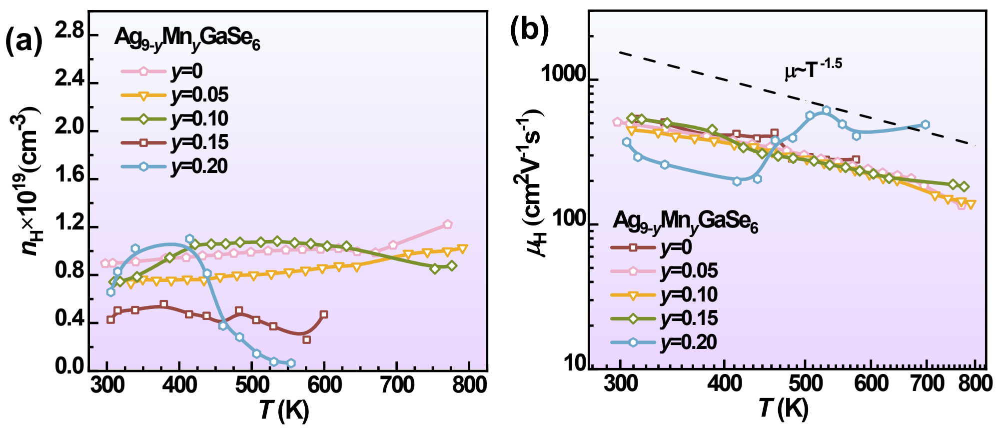

Figure S5. Temperature dependent (a) Hall carrier concentration and (b) Hall mobility for $Ag_{9-y}Mn_yGaSe_6$ (0≤y≤0.20) samples.

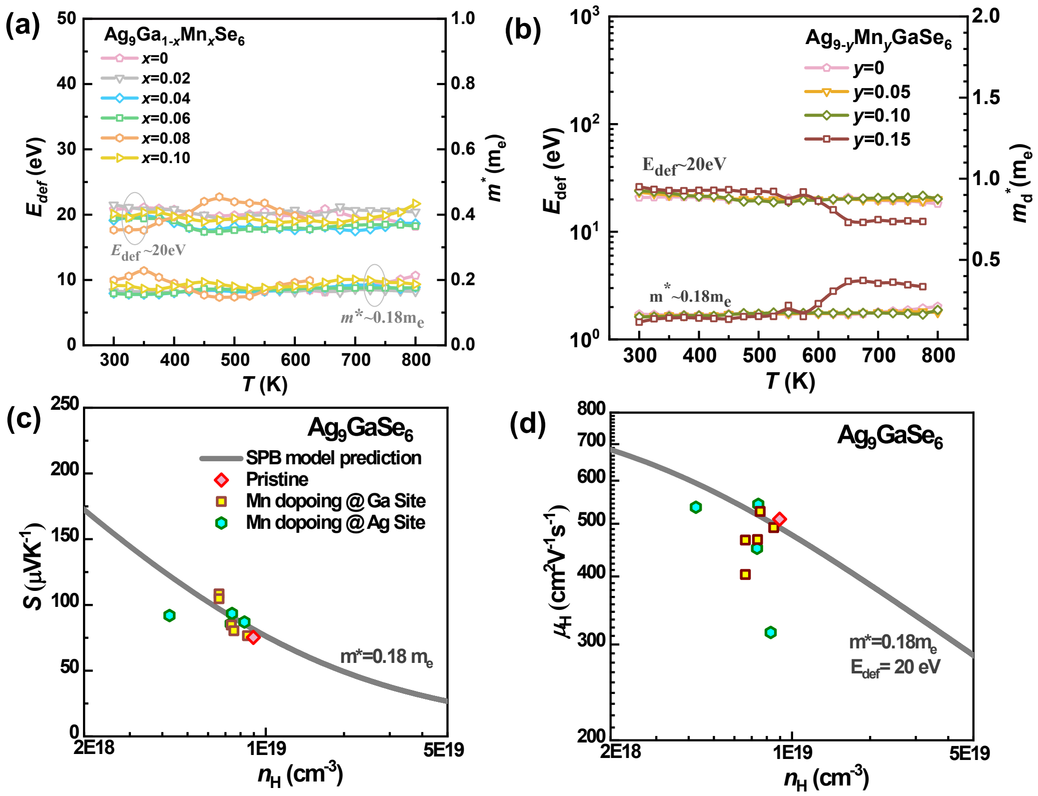

Figure S6. Temperature dependent deformation potential coefficient E_{def} and density-of-state effective mass m_d^* for (a) $Ag_9Ga_{1-x}Mn_xSe_6$ and (b) $Ag_{9-y}Mn_yGaSe_6$ samples. Hall carrier concentration dependent (c) Seebeck coefficient and (d) Hall mobility for Mn-doped Ag_9GaSe_6 samples.

Table S1. The hardness, elastic modulus and room temperature lattice thermal conductivity for typical thermoelectric materials.

Materials	Hardness (GPa)	Elastic modulus (GPa)	κ_L (W/m-K)	References
Ag_9GaSe_6	1.65	21.8	0.2	This work
MgAgSb	3.3	55	0.75	1, 2
$Si_{80}Ge_{20}$ alloy	14.5	147	4.71	3, 4
$Mg_{3.2}Sb_{1.5}Bi_{0.5}$	1.05	44.4	0.95	5
Half-heusler $Hf_{0.44}Zr_{0.44}Ti_{0.12}CoSb_{0.8}Sn_{0.2}$	13.9	231	2.45	6, 7
Half-heusler $Hf_{0.25}Zr_{0.75}NiSn$	9.9	213.9	4.24	6, 8

$Bi_{0.4}Sb_{1.6}Te_3$	1.5	48.7	0.76	6, 9
$Bi_2Te_{2.7}Se_{0.3}$	1.4	42.8	0.73	6, 10
Skutterudites $Yb_{0.35}Co_4Sb_{12}$	7.1	152.2	1.9	6
PbSe	0.7	74.1	1.7	6, 11
PbTe	3.7	26.6	1.81	12, 13
GeTe	6.9	119.4	1.0	12, 14
SnTe	1.89	51.1	1.93	15

Table S2. The measured transverse (v_t) and longitudinal (v_l) sound velocities, and the estimated mean (v_s) sound velocities, bulk modules (B), shear modules (G), Young's modulus (E), Grüneisen parameter (γ) for $Ag_9Ga_{1-x}Mn_xSe_6$ ($0 \leq x \leq 0.10$) and $Ag_{9-y}Mn_yGaSe_6$ ($0 \leq y \leq 0.20$) samples.

Mn-doped Ag_9GaSe_6	v_T **(m/s)**	v_L **(m/s)**	v_S **(m/s)**	B **(GPa)**	G **(Gpa)**	E **(GPa)**	γ
x=0.02	1128	2714	1276	39.8	8.9	24.9	2.57
x=0.04	1111	2656	1256	37.9	8.7	24.1	2.56
x=0.06	1140	2643	1288	36.8	9.1	25.2	2.47
x=0.08	1145	2734	1295	40.49	9.27	25.8	2.55
x=0.10	1120	2762	1268	41.8	8.8	24.6	2.64
y=0.05	1120	2741	1268	41.53	8.93	25.00	2.62
y=0.10	1123	2771	1271	42.64	8.97	25.17	2.65
y=0.15	1127	2864	1277	46.21	9.04	25.47	2.73
y=0.20	1124	2810	1273	44.16	9.00	25.27	2.68

Calculated elastic modulus for all the samples

The transverse (v_t) and longitudinal (v_l) sound velocities are measured to estimate the elastic modulus for all the $Ag_9Ga_{1-x}Mn_xSe_6$ ($0 \leq x \leq 0.10$) samples. The equations are given as below, where ρ is the density[16-18]:

$$v_s = \left[\frac{3}{2\left(\frac{1}{v_t}\right)^3 + \left(\frac{1}{v_l}\right)^3}\right]^{1/3} \quad (1)$$

Shear modules:

$$G = {v_t}^2 * \rho \quad (2)$$

Bulk modules:

$$B = {v_l}^2 * \rho - \frac{4}{3}G \quad (3)$$

Young's Elastic modules:

$$E = \frac{9BG}{3B + G} = \frac{\rho {v_t}^2(3{v_l}^2 - 4{v_t}^2)}{(3{v_t}^2 - 4{v_l}^2)} \quad (4)$$

Grüeneisen parameter:

$$\gamma = \frac{3}{2} * \frac{3{v_l}^2 - 4{v_t}^2}{{v_l}^2 + 2{v_t}^2} \quad (5)$$

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