SUPPORTING INFORMATION

1. XRD
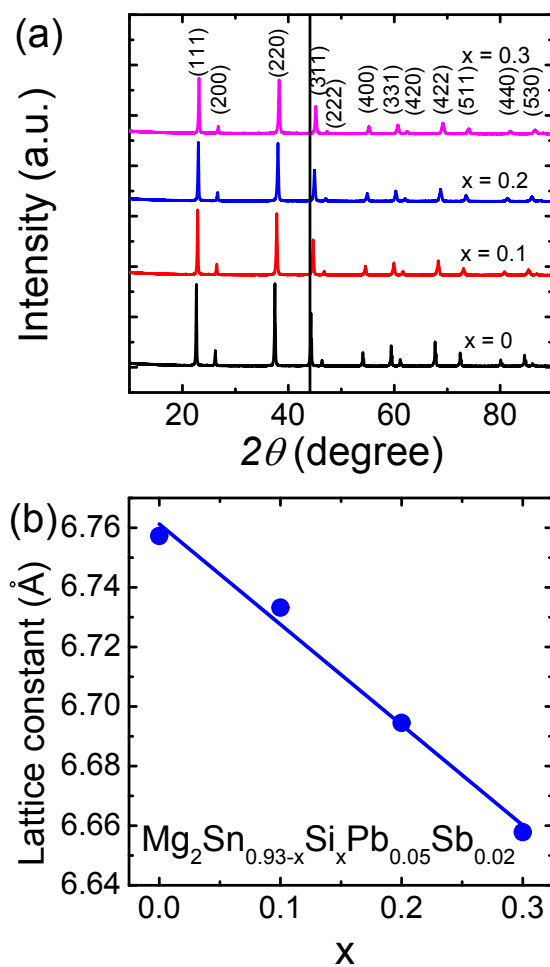

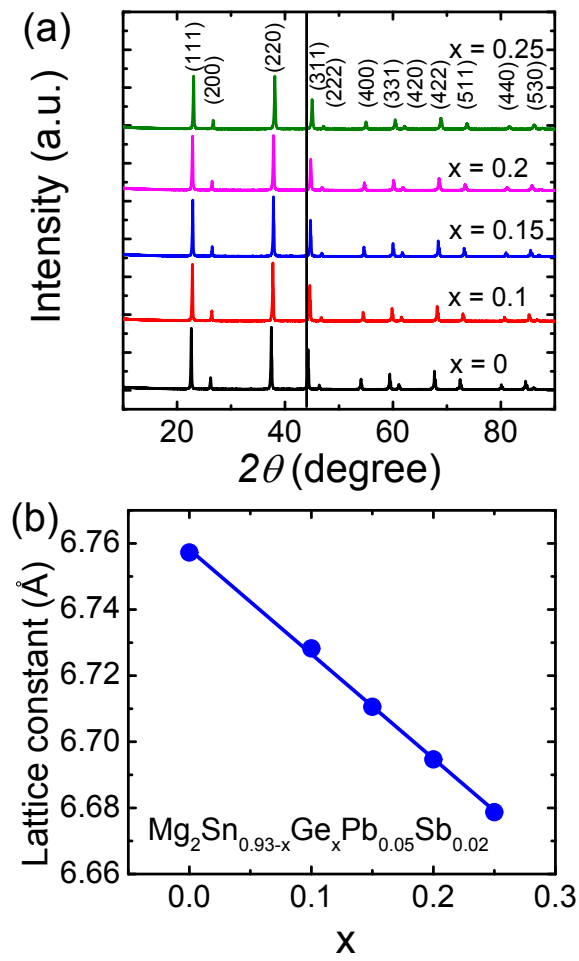Figure S1. (a) XRD patterns of hot pressed $Mg_2Sn_{0.93-x}Si_xPb_{0.05}Sb_{0.02}$. (b) Dependence of lattice parameter on composition of $Mg_2Sn_{0.93-x}Si_xPb_{0.05}Sb_{0.02}$.

Figure S2. (a) XRD patterns of hot pressed $Mg_2Sn_{0.93-x}Ge_xPb_{0.05}Sb_{0.02}$. (b) Dependence of lattice constant on the composition of $Mg_2Sn_{0.93-x}Ge_xPb_{0.05}Sb_{0.02}$.

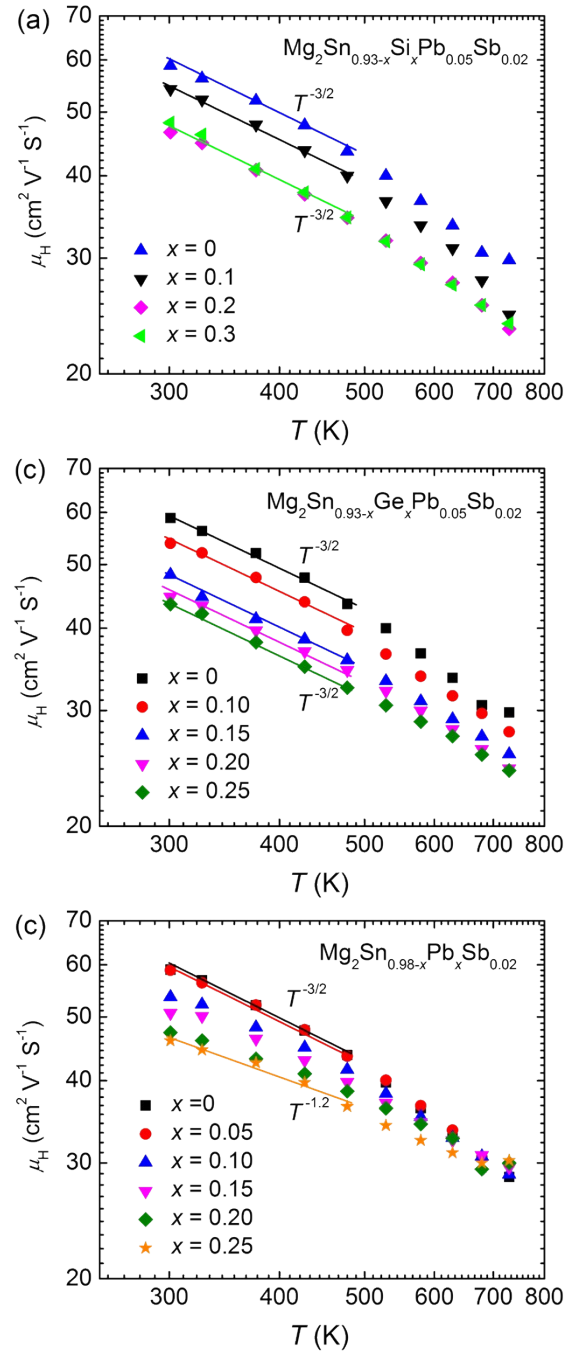## 2. Temperature dependent Hall mobility of Mg$_2$Sn based solid solutions

Figure S3. Temperature dependence of Hall mobility of Mg$_2$Sn based solid solutions. (a) Mg$_2$Sn-Mg$_2$Si-Mg$_2$Pb, (b) Mg$_2$Sn-Mg$_2$Ge-Mg$_2$Pb, and (c) Mg$_2$Sn-Mg$_2$Pb.

In order to clarify the carrier scattering mechanism, temperature dependent Hall mobility were calculated by using the measured electrical resistivity and room

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temperature Hall carrier concentration. Here, the assumption of constant Hall concentration is made and we are going to discuss it later when this assumption will be solid or be failed.

As shown in Fig. S3, it is clear that for Mg_2Sn-Mg_2Si-Mg_2Pb and Mg_2Sn-Mg_2Ge-Mg_2Pb solid solutions, the temperature dependence of Hall mobility for all the samples obey the $T^{-3/2}$ trend at lower temperature, which is exactly the acoustic phonon scattering mechanism[1]. This demonstrates that the alloying and ionization scattering do not significantly influence the scattering mechanism in Mg_2Sn-Mg_2Si-Mg_2Pb and Mg_2Sn-Mg_2Ge-Mg_2Pb solid solutions.

For the Mg_2Sn-Mg_2Pb solid solutions, the $T^{-3/2}$ trend can still be observed at lower concentration of Pb, however, deviation happens when the concentration of Pb is high. In this case we cannot simply claim that Pb-alloying will change the scattering mechanism of the carriers. The reason is that in the calculation of Hall mobility, it is assumed that Hall carrier concentration keeps constant in the whole temperature range, *i.e.*, bipolar conduction does not happen at higher temperature. This assumption can be regarded as solid for Mg_2Sn-Mg_2Si-Mg_2Pb and Mg_2Sn-Mg_2Ge-Mg_2Pb due to the suppressed bipolar conduction as we discussed in the manuscript. However, it is not true for the Mg_2Sn-Mg_2Pb solid solutions, especially when the concentration of Pb is high (due to enhanced bipolar conduction). Therefore, due to the excitation of minority carriers at higher temperature, the real Hall carrier concentration will be higher than the room temperature value, and hence the real Hall mobility will be lower than the calculated value. Thus, the deviation from the acoustic phonon scattering will not be as large as we observed. Similar to the Mg_2Sn-Mg_2Si-Mg_2Pb and Mg_2Sn-Mg_2Ge-Mg_2Pb solid solutions, it is reasonable to believe that the assumption of acoustic phonon scattering should still be held in the Mg_2Sn-Mg_2Pb solid solutions.

It should be pointed out that the temperature dependence of Hall mobility changes slightly in higher temperature for the Mg_2Sn based solid solutions. It is

presumably due to the inter-valley scattering or polar optical phonon scattering[2]. However, since this deviation is caused by the variation of temperature rather than the composition, therefore it will not influence the comparison of properties between the solid solutions.

3. Point defect scattering

$$\frac{\kappa_{L,\text{alloy}}}{\kappa_{L,pure}} = \frac{\arctan(u)}{u} \tag{S1}$$

$$u^2 = \frac{\pi^2 \theta_D \Omega}{h v^2} \kappa_{L,pure} \Gamma \tag{S2}$$

θ_D is the Debye temperature, its value is chosen as 400 K[1]. Ω is the volume per atom. h is the Planck's constant. v is the sound velocity which is taken as 4×10^5 cm s^{-1} [2]. Γ is the scattering parameter which was studied in detail by Klemens[3]. It contains three parts: mass difference, strain field and binding force difference. At here the binding force difference is not taken into consideration.

The mass difference scattering parameter Γ_M is given by:

$$\Gamma_{Mi} = x_i [(M_i - M)/M]^2 \tag{S3}$$

$$\Gamma_M = \sum_{i=1}^{n} \Gamma_{Mi} \tag{S4}$$

M_i is the atomic weight of ith substitution atom, M is atomic weight of the original atom and x_i is the ratio of ith substitution atom.

The strain field scattering parameter Γ_S is defined by [4]:

$$\Gamma s_i = x_i \{\varepsilon[(\delta - \delta_{i\text{sub}})/\delta]^2\} \tag{S5}$$

$$\Gamma_S = \sum_{i=1}^{n} \Gamma_{Si} \tag{S6}$$

$\delta_{i\text{sub}}$ is the atomic radius of the ith substitution atom, δ is the atomic radius of original atom. ε is a material related parameter and its value is taken as 23[5].

Therefore, the total scattering parameter Γ_{total} is:

$$\Gamma_{total} = \Gamma_M + \Gamma_S = \sum_{i=1}^{n} \Gamma_{Mi} + \sum_{i=1}^{n} \Gamma_{Si} \qquad\qquad (S7)$$
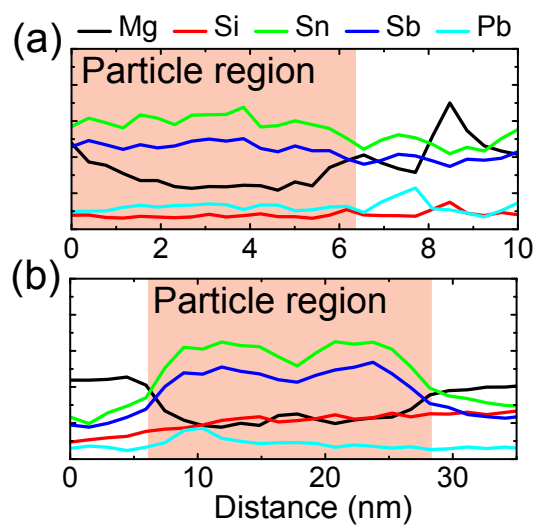
4. EDS

Figure S4. EDS results for the nanoparticles.

References

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