Supplementary Information for

Synergetic Enhancement of Thermoelectric Performances by Localized Carrier and Phonon Scattering in Cu_2Se with Incorporated Fullerene Nanoparticles

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Approximate interparticle distance of C_{60} nanoparticles by molar %

Table S1. Molar %, volumetric %, and estimated interparticle distance of embedded C_{60} nanoparticles

Molar %	Volumetric %	Interparticle distance (nm) (Assuming non-agglomerated C_{60})	Interparticle distance (nm) (Assuming agglomerated C_{60} similar with TEM result)
0.03	0.5	5.5	11.5
0.3	5	2.5	5.3
0.5	8	2.1	4.5
0.7	10	1.9	4.0

The relation between molar % and volumetric % in Table S1 can be expressed as follows,

$$V_{C_{60}} = \frac{Mol\%_{C_{60}} \times M_{C_{60}}}{\rho_{C_{60}}}, \; V_{Cu_2Se} = \frac{100\% \times M_{Cu_2Se}}{\rho_{Cu_2Se}}$$

$$Vol\% = \frac{V_{C_{60}}}{V_{Cu_2Se}} \times 100 \qquad \text{Eq. S1}$$

where V is the volume[cm^3], M is the molar mass[g mol^{-1}], and ρ is the density[g cm^{-3}]. To determine the amount of fullerene, we estimate the interparticle distance between fullerenes before the experiment. In our previous report[1], we found that the interparticle distance less than 15 nm was needed to reduce lattice thermal conductivity. To convert molar % to approximate interparticle distance in Table S1, we used the following equations.

$$\text{Number of } C_{60} : N_{C_{60}} = Mol\% \times N_A$$

$$\text{Volume of } Cu_2Se : V_{Cu_2Se} = \frac{100\ \% \times M_{Cu_2Se}}{\rho_{Cu_2Se}} \qquad \text{Eq. S2}$$

Then, the number density of C_{60} [count m^{-3}], η is,

$$\eta_{C_{60}} = \frac{N_{C_{60}}}{V_{Cu_2Se}} \quad \text{Eq. S3}$$

Assuming the fullerene nanoparticles are dispersed uniformly in a cubic matrix, then the line density of C_{60} [count m^{-1}], *η_line* is,

$$\eta\_line_{C_{60}} = \sqrt[3]{\eta_{C_{60}}} \quad \text{Eq. S4}$$

Finally, the interparticle spacing is the reciprocal of line density.

$$d_{spacing} = \frac{1}{\eta\_line_{C_{60}}} \quad \text{Eq. S5}$$

Hall measurement data

Table 2. Room temperature Hall measurement results

	n [cm^{-3}]	μ [$cm^2\ V^{-1}\ s^{-1}$]
pure Cu_2Se	4.70628	11.70039
0.03 mol%	1.83340	9.820827
0.3 mol%	1.85572	8.485708
0.5 mol%	1.74317	7.682623
0.7 mol%	2.20957	7.168936

Thermal conductivity calculation using the Callaway model

The thermal conductivities of the C_{60}/Cu_2Se nanocomposites are calculated using relaxation time approximation (Callaway model). The relaxation time is combined by Matthiessen's rule. (See Eq. (3) in main text) The relaxation time formula for intrinsic scattering process(Alloy, Normal, Umklapp) was discussed in our previous literatures.[2] For the formula of relaxation time for embedded nanoparticle is obtained from *W. Kim et al.*[3]

$$\frac{1}{\sigma_{NP}} = \frac{1}{\sigma_{Rayleigh}} + \frac{1}{\sigma_{near\ geometrical}} \quad \text{Eq. S6}$$

Here, both the scattering cross sections are described as,

$$\frac{\sigma_{Rayleigh}}{\pi R^2} = \chi^4 \left[\frac{\alpha^2}{4}\left(\frac{\Delta M}{M}\right)^2 + 3\alpha^8 \left(\frac{\Delta K}{K}\right)^2 \frac{\sin^4\left(\frac{\alpha\left|\vec{k}\right|\delta}{2}\right)}{\left(\frac{\alpha\left|\vec{k}\right|\delta}{2}\right)^4} \right] \times \left\{ \frac{\pi\left[\cos(4\chi) - 1 + (4\chi)\sin(4\chi) + 32\chi^4 - 8\chi^2\right]}{16\chi^6} \right\} \quad \text{Eq. S7}$$

$$\frac{\sigma_{near\ geometrical}}{\pi R^2} = 2\left\{ 1 - \frac{\sin\left[2\chi\left(\frac{k'}{k} - 1\right)\right]}{\chi\left(\frac{k'}{k} - 1\right)} + \frac{\sin^2\left[\chi\left(\frac{k'}{k} - 1\right)\right]}{\left[\chi\left(\frac{k'}{k} - 1\right)\right]^2} \right\} \quad \text{Eq. S8}$$

where, the M is atomic mass, K is spring constant, α is trigonometric ratio, δ is interparticle distance, k is wavevector, and χ is size parameter. Using Eq. S6, the relaxation time for phonon scattering by embedded C_{60} is,

$$\tau_D = \frac{1}{\eta_{C_{60}} v \sigma_{NP}} \quad \text{Eq. S9}$$

here, number density of C_{60} is obtained by Eq. S3, v is phonon group velocity.

Reference

1 Hongchao Wang, Je-Hyeong Bahk, Chanyoung Kang, Junphil Hwang, Kangmin Kim, Jungwon Kim, Peter Burke, John E. Bowers, Arthur C. Gossard, Ali Shakouri, and Woochul Kim, Proceedings of the National Academy of Sciences of the United States of America **111** (30), 10949 (2014).

2 J. Hwang, H. Kim, M. K. Han, J. Hong, J. H. Shim, J. Y. Tak, Y. S. Lim, Y. Jin, J. Kim, H. Park, D. K. Lee, J. H. Bahk, S. J. Kim, and W. Kim, Acs Nano **13** (7), 8347 (2019).

3 W. Kim and A. Majumdar, Journal of Applied Physics **99** (8), 084306 (2006).