

Supplementary material

Theoretical insights into lattice thermal conductivity and thermal expansion of CoNiFe medium-entropy alloys

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Supplementary method

In this paper, we adopt the First Avalanche (FA) method,

$$F(t) = \frac{|\sigma(\text{Cor}(t))|}{|E(\text{Cor}(t))|} \quad (\text{S1})$$

where σ and E are the standard deviations and average value in a small period, respectively. In this paper, we choose the moment when $F(t)$ suddenly increases and jumps as the cutoff time. In principle, there is no finite size effect in the EMD method [1]. However, as the simulated supercells increase, low-frequency phonons near the Γ -point are excited. These trends make an important contribution to the thermal conductivity [2]. In addition, there is an increased likelihood of phonon-phonon scattering, which decreases the thermal conductivity. These two competing factors result in the EMD method still having finite size effects. Therefore, the effect of supercell size on thermal conductivity must be taken into account in simulations [3, 4].

The heat current autocorrelation calculations are performed for 10 times with an autocorrelation time of 100 ps. A typical average heat current autocorrelation in a 5×5 supercell is shown in Fig. S1 (a). To determine the cutoff time, we further calculate $F(t)$, as shown in Fig. S1 (b), and choose the moment when $F(t)$ suddenly increases and jumps as the cutoff time. To ensure the accuracy of the calculation, we calculate the cutoff time for different MEAs. The results of the lattice thermal conductivity (Fig. S2) indicate that, there is a clear finite-size effect, with the thermal conductivity converging to 5.05 ± 0.20 W/mK when $N \geq 3$. This agrees well with the lattice thermal conductivity (1.95-7.83 W/mK) reported in previous experiments [5, 6].

To evaluate the thermal expansion, the systems are heated to different temperatures (400-1500 K) in the NPT ensemble after relaxation in NPT and NVT at 300 K. We calculate the volume expansion coefficient (α_V) through the following equation,

$$\alpha_V = \frac{dV}{VdT} \quad (\text{S2})$$

where V is the volume, and T is the temperature. We usually refer to the coefficient of thermal expansion (α) as the coefficient of linear expansion, which can be calculated from the coefficient of volume expansion,

$$dV = d(hwl) = wldh + hldw + hwdl = hwl \left(\frac{dh}{h} + \frac{dw}{w} + \frac{dl}{l} \right) \quad (\text{S3})$$

$$\alpha = \frac{dL}{LdT} = \frac{1}{3} \frac{dV}{VdT} = \frac{1}{3} \alpha_V \quad (\text{S4})$$

where h , w , and l are the lengths of the systems in the x , y , and z directions, respectively.

Supplementary figure

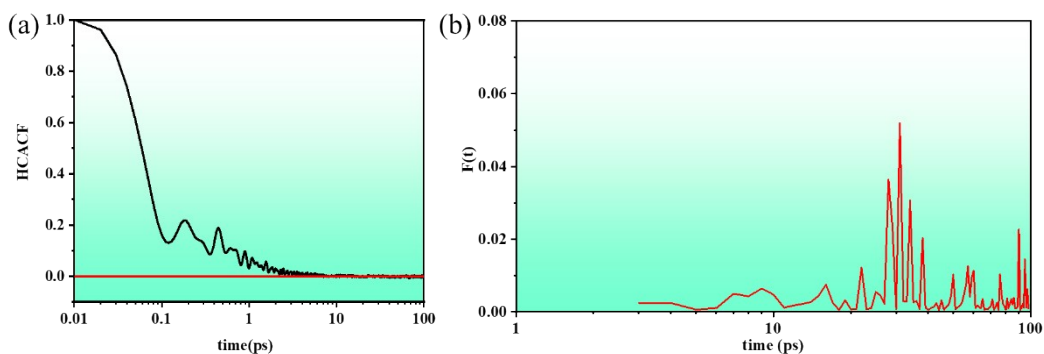


Fig. S1. (a) The normalized HCACF in a $5 \times 5 \times 5$ MEA supercell (other simulations are similar). (b) $F(t)$ in a $5 \times 5 \times 5$ MEA supercell (other simulations are similar).

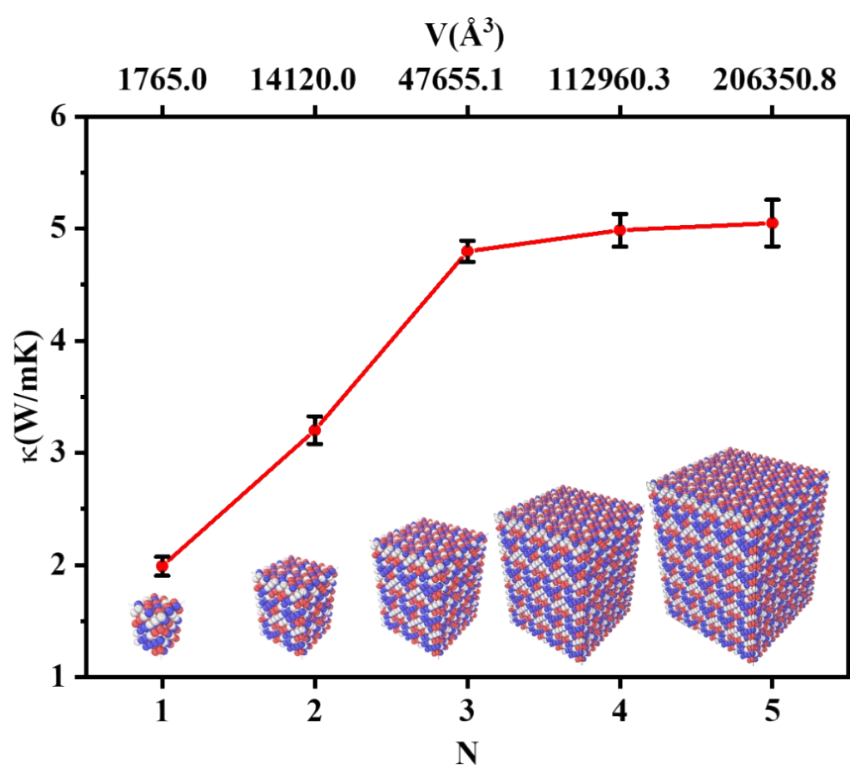


Fig. S2 The lattice thermal conductivity of the CoNiFe MEA at 300 K versus supercell size ($N \times N \times N$ unit cells).

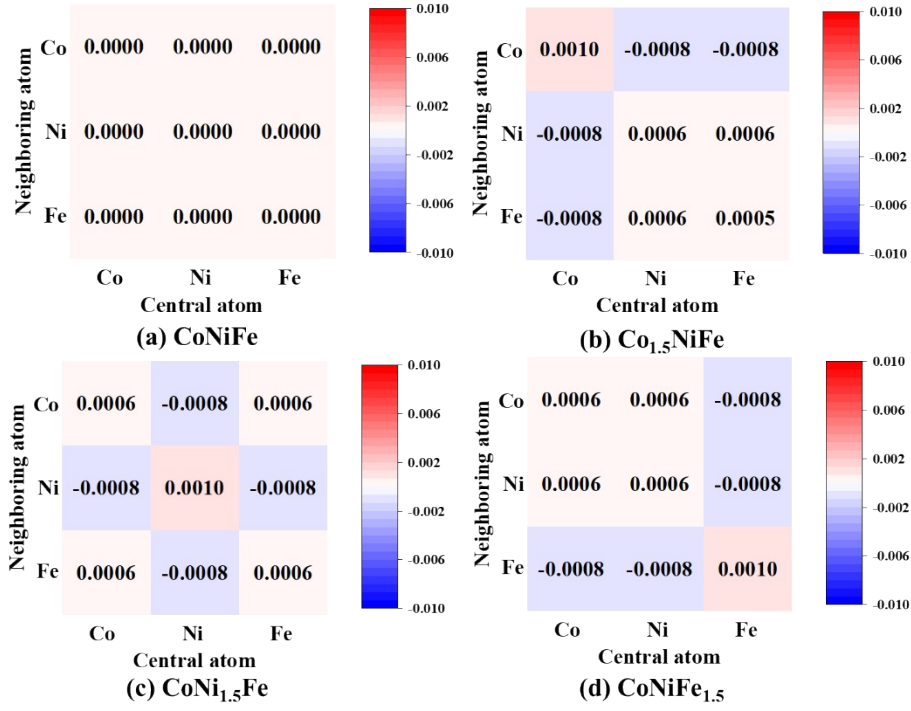


Fig. S3. The WCF of different MEAs.

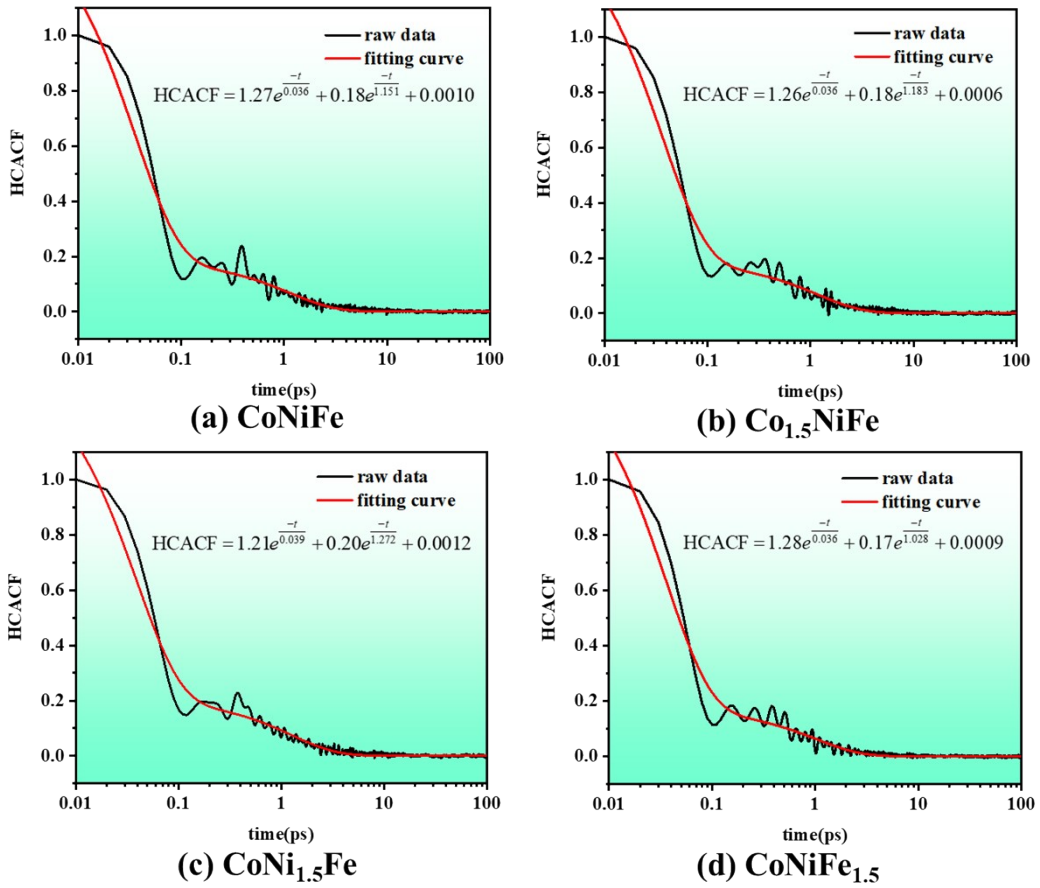


Fig. S4. The double Exponential Fitting of HCACF for different MEAs.

Supplementary references

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