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Thermal Rectification of Sierpiński Tetrahedron Fractals Assembled from Supertetrahedral T₂-type Tin Selenide Clusters

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Abstract

To explore the potential of atomic clusters in device applications, we illustrate the thermal rectification (TR) effect and underlying mechanisms in the cluster-assembled Sierpiński tetrahedron (ST) fractals. Using the synthesized supertetrahedral T₂-type Sn₄Se₁₀ clusters as building blocks and nonequilibrium molecular dynamics with a machine-learning neuroevolution potential trained on a multidimensional *ab initio* dataset, we show that TR coefficient of the assembled ST₄ fractal reaches 17.4% at an average temperature of 300 K with a temperature bias of 20 K. This further increases to 39.4% under the same temperature condition for the higher-level fractal ST₅ as the void ratio and surface roughness increase significantly with the fractal level. Moreover, unlike in conventional atom-based nanosystems, the dependence of TR coefficient on temperature bias in the ST fractals is nonmonotonic, where a high TR efficiency can be achieved just with a small temperature bias. Our work demonstrates the merits of cluster-assembled fractals for high-performance thermal rectifiers and the advantages of cluster-assembled materials for thermal energy management.

As an electric diode-like effect for active control over heat flow, thermal rectification (TR) enables heat flux to change asymmetrically as the temperature bias is reversed [1,2], exhibiting broad applications in phononic devices [3] including thermal diodes [4], thermal transistors [5], thermal logic gates [6], thermal memory [7], and thermal energy conservation and storage [8]. The TR efficiency depends on the difference in thermal conductivity (κ) between two opposite directions and is measured with the TR coefficient defined as $\eta = (\kappa_f - \kappa_b)/\kappa_b \times 100\%$, where f and b represent the forward and backward heat flows, respectively.

TR effect arises from the interplay between structural gradient and lattice anharmonicity [9,10] induced by either designing asymmetric structures, applying strain, introducing defects or dopants, or forming heterojunctions [11]. Significant TR is expected to exist in the asymmetric nanostructures. However, the currently reported systems are mainly restricted to atom-based substances with simple geometries such as carbon nanotubes, silicon nanowires, and graphene-based nanostructures like trapezoid or triangular patches, branched nanoribbons, and multilayer junctions [10,12-15]. Moving beyond conventional atom-based systems, cluster-assembled polyhedra have recently been proposed as thermal rectifiers [16]. Since cluster assembly offers a flexible strategy for modulating spatial configurations in TR, a pertinent question arises: Are there other unconventional methods for assembling clusters?

Fractals are well-known for their rough surfaces or edges with self-similarity at different scales [17]. Consequently, they are endowed with inherent nonlinearity [18], holding potential as a natural platform for TR, which could be constructed from clusters acting as flexible building blocks for designing new functional materials [19,20]. Especially, the vibrational spectra of cluster-assembled materials are susceptible to significant changes due to the coupling of intracluster and intercluster vibrations. Motivated by the experimental synthesis of supertetrahedral T₂-type tin selenide clusters [21], here we propose novel cluster-based thermal rectifiers using the cluster-assembled Sierpiński tetrahedron fractals, and explore the resulting TR behaviors and microscopic mechanisms by employing machine-learning molecular dynamics.

Math model of Sierpiński tetrahedron fractals for thermal rectification

As the three-dimensional analog of the Sierpiński triangle [22], the Sierpiński tetrahedron (ST) is a fractal with tetrahedron skeletons (Fig. 1 and Fig. 2a). They exhibit self-similarity at different scales, which can be formed by assembling regular tetrahedrons. Taking the second generation of ST fractal (ST₂ in Fig. 1) as an example, there are totally 16 basic regular tetrahedrons, which are identical and arranged in the same orientation with the sequence of (1, 3, 3, 9) from the top vertex to the bottom base, displaying highly hierarchical asymmetry in shape that the tetrahedral unit itself is geometrically asymmetric and such asymmetry is extended to the dimension of the whole ST fractal.

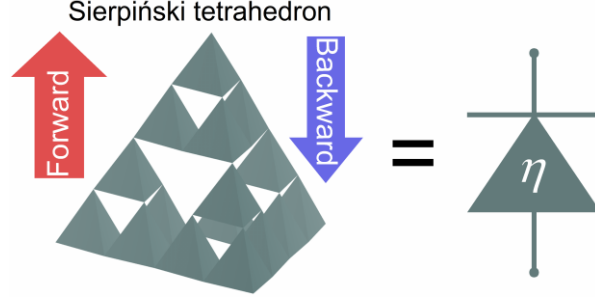


FIG. 1. Model of thermal rectifiers using Sierpiński tetrahedron fractals. The arrows represent forward and backward heat flows, respectively; the triangle symbol represents a thermal diode in thermal circuits.

To quantitatively illustrate the thermal rectification (TR) effect using the ST fractals as thermal rectifiers (Fig. 1), we introduce the generalized heat flux transmission function (τ) [23] as

$$\tau_{f/b} = \frac{J_{f/b}}{k_B \Delta T}$$

where J_f (J_b) is the forward (backward) heat current from the bottom base to the top vertex (reversely), k_B is the Boltzmann constant, and ΔT is the temperature difference. With the definition of thermal conductivity $\kappa = J/(S \Delta T/L)$, we can obtain the ratio between forward and backward κ ,

$$\frac{\kappa_f}{\kappa_b} = \frac{\tau_f}{\tau_b}$$

From a low-generation ST_{n-1} to the high-generation ST_n (n labeling the fractal level), the ST fractal architecture can be treated as an in-parallel-series thermal circuit. By making a very rough but intuitive assumption that four ST_{n-1} are independent and do not interact with each other, we finally derive the ratio of forward and backward overall κ for ST_n ,

$$\frac{\kappa_f^{ST_n}}{\kappa_b^{ST_n}} = \frac{\tau_f^{ST_n}}{\tau_b^{ST_n}} = \frac{\tau_f^{ST_{n-1}} \cdot \tau_f^{ST_{n-1}}}{\tau_b^{ST_{n-1}} \cdot \tau_b^{ST_{n-1}}} = \left(\frac{\kappa_f^{ST_{n-1}}}{\kappa_b^{ST_{n-1}}} \right)^2 \quad (1)$$

This formula unveils the scalability of the rectifying function of thermal rectifiers using the ST fractals with different fractal levels, namely, the difference in κ between forward and backward directions is enhanced in a higher fractal level. Based on these mathematic relations, one sees that the ST fractals could be a natural amplifier for the TR effect.

Materials realization of Sierpiński tetrahedron fractals

The high ratio of voids in the ST fractals makes it difficult to realize them in the traditional material systems with atoms as building units. The paradigm shift from elemental atoms to clusters opens another route, where the fractal architecture can be constructed with atomically precise clusters through a bottom-up assembling approach. In fact, several fractal patterns have been synthesized by molecular assembly on 2D surfaces, such as Sierpiński triangle or hexagon gasket through halogen bonds [24], artificial manipulation [25], coordination interaction [26,27], and covalent bonds [28]. Similarly, the ST fractals could be achieved via the assembly of tetrahedron-shaped clusters, among which metal chalcogenide supertetrahedral clusters (MCSCs) are of special interest for their uniform sizes and well-defined structures synthesized in experiments [21,29]. Especially, MCSCs consisting of basic metal-chalcogen coordination tetrahedrons can serve as secondary building units in porous frameworks to form a hierarchical assembly.

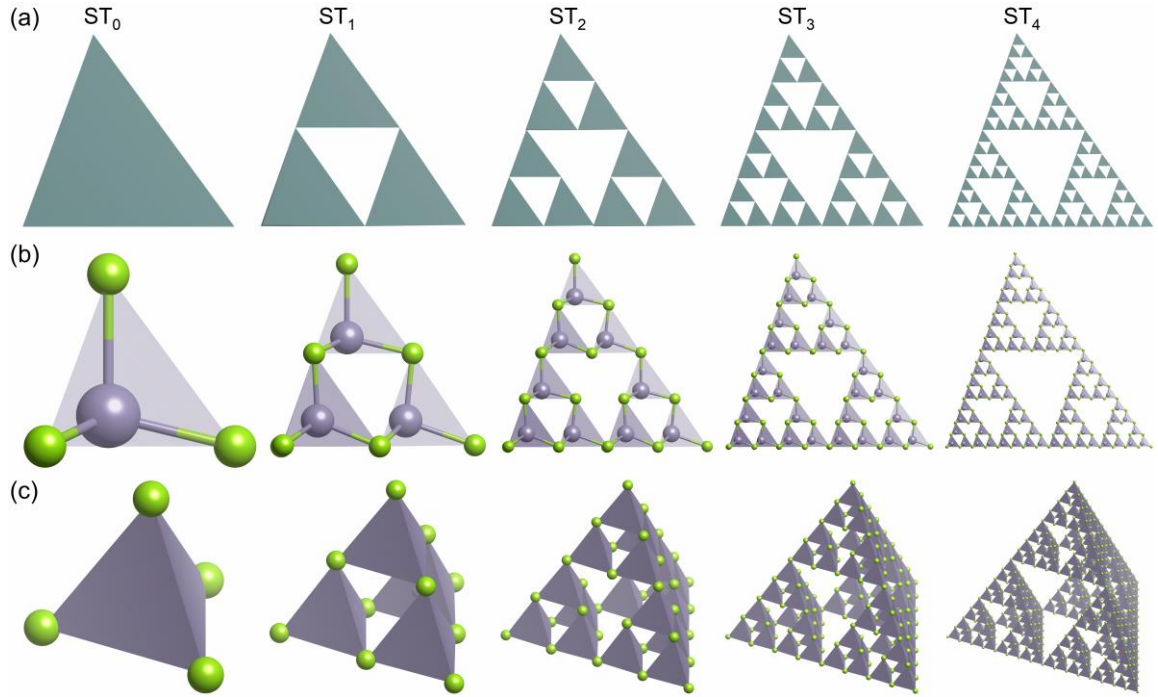


FIG. 2. Materials realization of Sierpiński tetrahedron fractals via the assembly of supertetrahedral T₂-SnSe clusters. (a) Fractal geometry models from the zeroth (ST₀) to fourth (ST₄) generations; (b) side views and (c) perspective views of the corresponding cluster-assembled frameworks.

To this end, we propose a possible materials realization of the ST fractals via the assembly of supertetrahedral T₂-type Sn₄Se₁₀ clusters (denoted as T₂-SnSe). By sharing

the corner bicoordinated Se (μ_2 -Se) atoms, the connection mode widely observed in MCSC-based frameworks [21], we then obtain a series of ST fractals, ST_n (Fig. 2b,c), where each generation is from the hierarchical fusion of the supertetrahedral T_2 -SnSe clusters. The zeroth generation of this ST_n series, ST_0 , is the basic SnSe₄ tetrahedron. For the first-generation ST_1 , it is the supertetrahedral T_2 -SnSe cluster itself that contains four SnSe₄ tetrahedrons. As to the next generation, ST_2 can be regarded as a super-supertetrahedral cluster with four supertetrahedral T_2 clusters ($T_{2,2}$, a special type of MCSCs as reported experimentally [30]). During the iterative assembling, all the Se atoms at the connection sites between clusters keep the same bonding feature of angular bicoordination as that within the T_2 -SnSe cluster units.

Machine-learning neuroevolution potential

To model the potential energy surface (PES) and set the ST structures with proper geometric parameters for TR simulation, stable T_2 -SnSe cluster-assembled frameworks containing a similar bonding environment as the ST fractals are desired. In the ST_n at different fractal levels, the overall coordination environment simply includes the tetracoordinated Sn with four Se atoms and the angular bicoordinated Se with two Sn atoms for both intracluster and intercluster linkage, except for four end Se atoms at the vertices of ST. According to these features, we assemble the supertetrahedral T_2 -SnSe clusters into stable 2D and 3D crystalline frameworks also by sharing the corner μ_2 -Se atoms as shown in Fig. S1-S3 in the Supplemental Material (SM) [31] (see, also, references [8,32-37] therein).

Then, we develop a machine learning model—neuroevolution potential (NEP) [38,39] on a multidimensional density functional theory (DFT) dataset through enhanced sampling configuration space from the long-time *ab initio* molecular dynamics trajectories up to 0.1 ns as illustrated in Fig. 3a and elaborated in SM. The evolution of various loss functions as a function of training generations is plotted in Fig. 3b. According to the root-mean-square error (*RMSE*) and correlation coefficient (R^2) on the test set (Fig. 3c and Fig. S4), one sees that the NEP model is well-trained and shows good agreement with DFT results for energy, force, and virial. Also, the phonon dispersions calculated with the NEP potential match well with those from DFT (Fig. S5). Therefore, it is reliable to employ the NEP potential to describe PES with DFT accuracy for performing large-scale molecular dynamics. For example, the thermal stability is simulated with NEP for the ST fractal structures (Fig. S6).

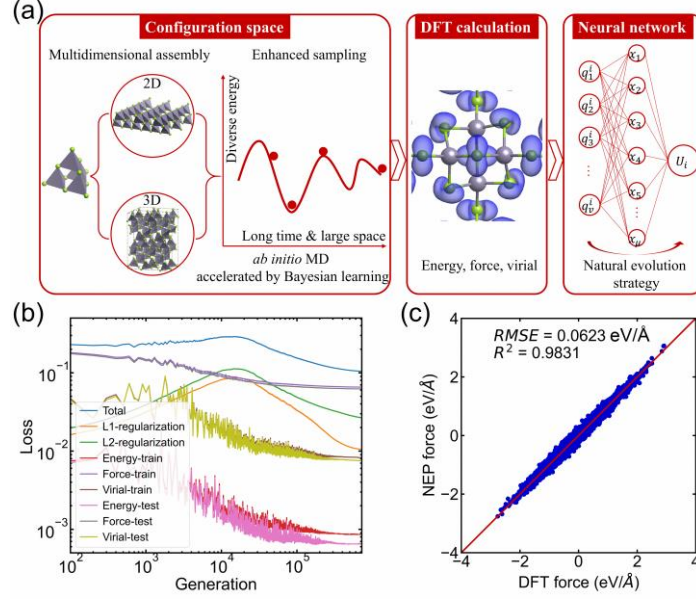


FIG. 3. Machine-learning NEP potential for the T_2 -SnSe cluster-assembled structures. (a) Development workflow; (b) evolution of loss functions during the training process; (c) forces predicted from NEP in comparison with the test data from DFT calculations.

Thermal rectification efficiency

By imposing a temperature bias between the top vertex and bottom base of the ST_4 fractal (Fig. S7), the resulting heat currents flow along a three-fold axis (the forward direction is from the bottom base to the top vertex while the backward is reverse, as shown in the inset in Fig. 4a). Significant TR effect occurs in ST_4 as observed from the heat current differences between the two directions (Fig. 4a), and the TR coefficient (η) reaches 17.4% at the average temperature $T_{ave} = 300$ K with a temperature difference $\Delta T = 20$ K (Fig. 4c). The favored direction of heat conduction is from the wider base to the narrower vertex, consistent with the results found in triangular or trapezoid graphene nanoribbons [10,13], where heat transfer from wide to narrow is more favorable than the opposite direction.

To figure out the underlying origin of the TR effect in ST_4 fractal, we compare the vibrational density of states (VDOS) $D(\omega)$ in heat conduction along the two directions (Fig. 4e), which exhibits distinct differences in the state distribution across the frequency domain of ST_4 . Note that the thermal conductivity $\kappa(\omega)$ is directly related to $D(\omega)$: $\kappa(\omega) \propto v_g(\omega) \lambda(\omega) \hbar \omega D(\omega) \partial[f(\omega, T)]/\partial T$, where λ and f are mean free paths and the distribution function, respectively. As a result, the discrepancy of mode-resolved thermal conductance exists in ST_4 (Fig. 4f), where the contribution from low-frequency modes (as the main heat carriers) in the forward flow

is larger than the backward, leading to the higher κ_f . Such differences in ST4 arise from the asymmetric restriction on vibrational states in the lateral (xy) dimensions [8], which makes local DOS (thus κ) not only depends on the temperature (Fig. S8) but also the region width (Fig. S9).

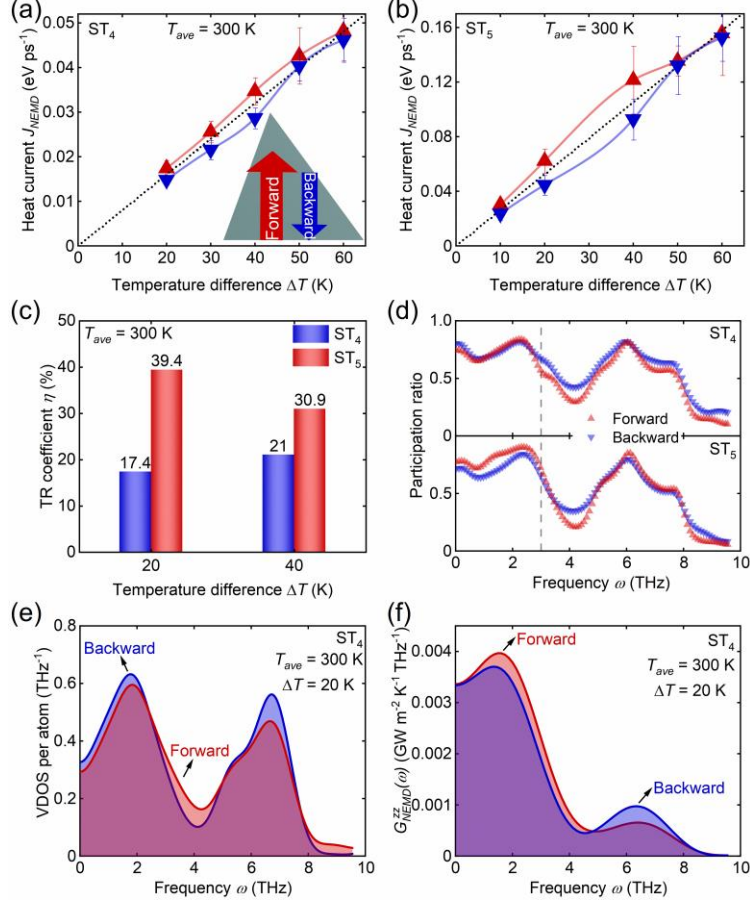


FIG. 4. Thermal rectification of the Sierpiński tetrahedron fractals assembled from the supertetrahedral T₂-SnSe clusters. (a, b) Heat current versus temperature bias, and dotted lines for linear Fourier's law, (c) thermal rectification coefficients, and (d) participation ratio of vibrational states for ST4 and ST5; (e) vibrational density of states and (f) spectral thermal conductance in ST4.

In addition, from the temperature profile along the heat flow (Fig. S10), we observe that the temperature variation with respect to spatial positions is highly nonlinear in ST4, which is induced by both the asymmetric geometry and high interior porosity. Particularly, these abundant voids are intrinsically and hierarchically asymmetric, very different from the extrinsically introduced asymmetric dopants or defects in conventional atom-based systems [12,40]. Accordingly, the temperature gradient within ST4 as a function of spatial positions varies from the wider base to the narrower vertex.

Moreover, nonlinear heat transfer exists in ST₄ (Fig. 4a), where the heat current deviates from the linear relation with the temperature difference (gradient) between thermal reservoirs, namely, thermal conductivity κ depends on the temperature gradient. In contrast to the conventional atom-based systems, such nonlinear heat transfer in the fractals benefits from the synergy of both the hierarchical assembly of cluster units and the fractal geometry. Based on these complex dependences of κ on temperature, width, and temperature gradient, each as a function of space positions, the κ in ST₄ fractal thus exhibits strong and nonlinear dependence on space location. Then, due to the correlated dependence of κ on temperature and space, the structural asymmetry in space will induce nonreciprocal heat conduction as discussed in SM for details.

To numerically examine the amplifying effect of the ST fractals on TR as deduced in Eq. (1), comparisons are made between ST₄ and ST₅ fractals. We show that η of ST₅ increases to 39.4% at $T_{ave} = 300$ K with $\Delta T = 20$ K (Fig. 4c), which can be attributed to the unique feature of fractal as shown in Fig. 2, where the void ratio increases exponentially with the increase of fractal level n . Thus, ST₅ fractal has significant interior and exterior surface roughness, which results in more localized vibrational modes (Fig. 4d) measured with the participation ratio defined as [41]

$$PR(\omega) = \frac{1}{N} \frac{(\sum_i D_i^2(\omega))^2}{\sum_i D_i^4(\omega)}$$

where $D_i(\omega)$ is vibrational density of states on atom i and N is the total number of atoms. The PR value varies between $O(1)$ for completely delocalized states and $O(1/N)$ for completely localized ones. In particular, the low-frequency states (< 3 THz) in the backward heat flow are more localized than the forward ones in ST₅, leading to a larger difference in the thermal transport capacity between forward and backward directions therein (details in Fig. S11). Meanwhile, the nonlinear heat transfer is further enhanced in ST₅ (Fig. 4a vs. Fig. 4b), i.e., a stronger dependence of κ on temperature gradient, resulting in a more inseparable dependence of κ on temperature and space, thus promoting TR efficiency. Hence, we inspect the variation of local thermal conductance with spatial regions in the flow direction (Fig. S12), where the narrow region near the top vertex dominates the difference between the forward and backward heat conduction, demonstrating the significance of the lateral confinement of vibrational modes in the nanostructures. In this regard, the narrow region in a higher-level fractal with higher-ratio voids plays a critical role in enhancing TR efficiency.

It is worth noting that the dependence of η on ΔT is not monotonic (Fig. S13). As ΔT becomes larger, the η first increases and then decreases, different from the

monotonic increase in atom-based compact systems [8,11]. When inspecting the nonlinear relation of heat currents with temperature bias (Figs. 4a,b), one can find that the ratios of heat currents to ΔT (proportional to the magnitude of apparent κ) exhibit different behaviors with ΔT between forward and backward heat flows. This is related to the evolution of mode-resolved contribution: the contribution from low-frequency states to forward κ is reduced while that from high-frequency modes is enhanced as ΔT increases; whereas the variation is reversed in the backward case (Fig. S14). Such an unusual dependence of η on ΔT would lead to an optimal TR efficiency at a moderate temperature bias, e.g., η reaches 39.4% at $\Delta T = 20$ K for ST₅, which is much higher than the efficiency of the pristine graphene trapezoid with $\eta < 10\%$ [13,35]. Therefore, the ST fractals assembled from the supertetrahedral clusters can display better TR performance under a narrow thermal bias, making the application in thermal devices more effective.

In summary, going beyond the studies reported hitherto on clusters for hydrogen storage [42,43], CO₂ activation [44], batteries [45,46], and thermoelectricity [47], we explore the role of cluster-based materials for thermal rectifiers using the Sierpiński tetrahedron fractals assembled from the supertetrahedral T₂-SnSe clusters. The resulting cluster-assembled fractals have a high void ratio and surface roughness changing exponentially with the fractal levels, which makes the temperature vary with spatial positions highly nonlinearly and induces nonlinear heat transfer. Thus, the thermal conductivity therein exhibits high asymmetry, depending sensitively on the transport direction. Such cluster-assembled fractals provide a unique platform for designing high-performance thermal diodes used in phononics and thermal management.

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