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Supplemental Material

Negative differential thermal resistance through nanoscale solid-fluid-solid sandwich structures

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Section A: The Lennard-Jones parameters

TABLE S1 The Lennard-Jones parameters[1,2]

	σ (Å)	ε (K)
He-He	2.55	10
Ne-Ne	2.82	32
Ag-Ag	2.547	4000
Al-Al	2.551	4736

Section B: MD details of the sandwich structure

Random velocities that follow the Maxwell distribution corresponding to a constant temperature (equal to the left thermostat temperature T_L for SL, the right thermostat temperature T_R for SR, and (T_L +T_R)/2 for the fluid) are assigned to the solid atoms and fluid molecules as the initial conditions. Newton's equations of motion are integrated by the Velocity-Verlet algorithm with a time step Δt=1fs. The cutoff distance is 0.75 nm. The system is equilibrated in an *NVT* (*N* is the number of fluid molecules, *V* is the fluid volume, and *T* is the temperature) ensemble for 1 ns. Then, after a relaxation time of 0.5 ns, the Berendsen thermostats with T_L and T_R are applied to the two-layer atoms adjacent to the leftmost and rightmost layers, respectively. The simulation systems are allowed to relax under the thermostats for

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1.5 ns before the data are collected for 5 ns. The velocities of all the vibrating atoms in the left or right thermostats are scaled at each time step in order to maintain the temperatures at T_L or T_R. The difference between the total kinetic energy, E, of the vibrating atoms in thermostat W (W represents L or R) and the energy corresponding to the desired thermostat temperature T_W, $\Delta E = E - 3N_W k_B T_W/2$, are calculated at each step before the velocities are scaled, where N_W is the number of atoms in thermostat W and k_B is the Boltzmann constant. Then, the heat flux $q = <\Delta E>/(\Delta t \cdot S)$, where S is the cross-sectional area, and <•>stands for the temporal average.

Section C: The temperature distribution of the nanoconfined fluid system

In the direction of thermal transport (x direction), the fluid part F is divided into 10 bins, and each solid part (SL and SR) is divided into 3 bins. The temperature in the i-th bin, T_i, is calculated based on the peculiar velocities of the molecules in the bin relative to the streaming velocity of the bin,

$$T_i = \sum_{i=1}^{N_i} m\left(v_i - \bar{v}\right)^2 / 3N_i k_B \tag{S1}$$

where m is the mass of the molecule, v_i is the velocity of the molecule, N_i is the total molecule number in the bin, and $\bar{v}$ is the mean velocity of the molecules.

The temperature distributions of the nanoscale confined fluid systems with three cold surface temperatures (T_R=100K, T_R=180K, T_R=260K) are plotted in Fig.S1. Because the thermal resistances of the solid parts can be usually ignored compared to that of the fluid part, the temperature distribution in the solid parts is almost flat, as expected.

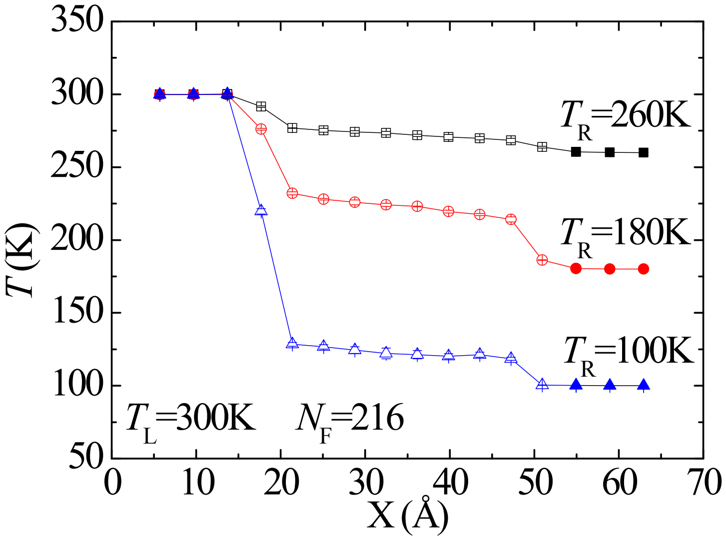

Fig.S1. Temperature distributions of the nanoscale confined fluid systems with three cold surface temperatures (T_R=100K, T_R=180K, T_R=260K). Solid and open symbols represent the solid and the fluid parts, respectively. Here, T_L=300K and N_F=216.

Section D: The mechanism of the adsorption at the cryogenic temperature

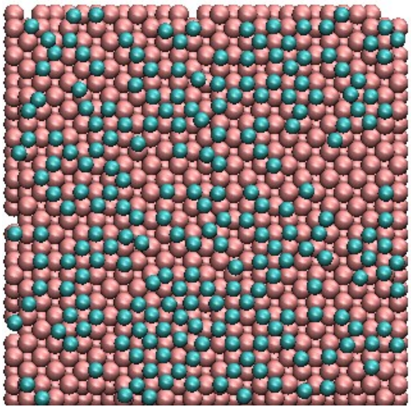

Fig.S2. Sideview for the adsorbed fluid molecules on the cold (right) surface. Here, T_L=300K, T_R=80K, and N_F=216.

The fluid molecules can be adsorbed on the cold surface because they cannot overcome the binding energy between the solid surface and the fluid. In our MD simulations, the binding energy ε_{SF}=217K. So, the adsorption can occur when T_F<217K, where T_F is the temperature of the fluid near the wall. In the case of T_R=80K, the fluid molecules are tightly adsorbed on the cold solid surface and a layer of ordered fluid molecules are formed in alignment with the solid atoms (see Fig.S2).

Section E: The heat transfer for a nanoconfined liquid

As mentioned in section III, the NDTR effect can be tuned by changing the number density of the confined fluid, N_F. With increasing N_F, the NDTR effect is weakening. If a liquid is employed instead of a gas, the effect of the molecule adsorption on the cold surface is weaken and there should be no NDTR effect in the present sandwiched structure, as shown in Fig. S3. With increasing N_F (for N_F=2188, the fluid is a liquid), the heat transfer is enhanced but the NDTR effect disappears and the heat flux is almost linearly proportional to ΔT.

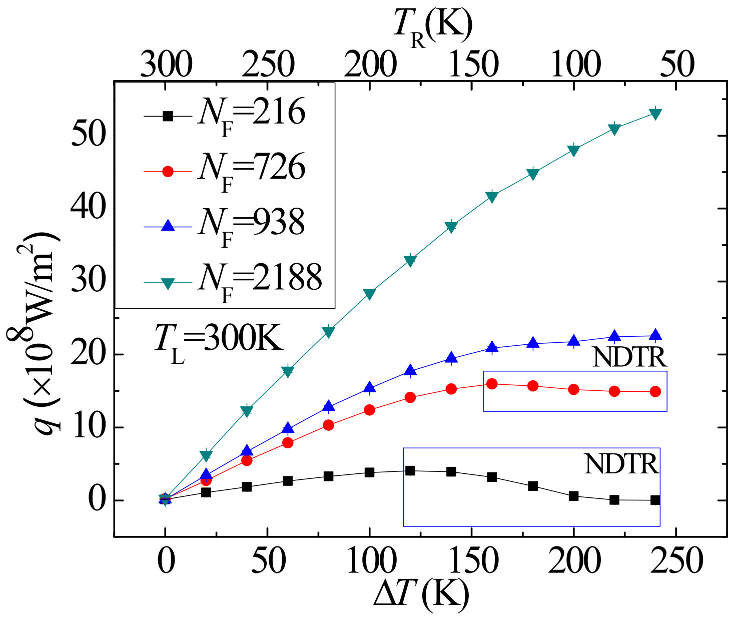

Fig.S3. The dependence of NDTR on the number density of the confined fluid.

References:

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