Supplementary Information for

Thermoelectricity in atom-sized junctions at room temperatures

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The Supplementary Information includes:

1. Supplementary Figures (Fig. S1-S10)

2. Supplementary references
Figure S1. Fabrication process flow of microheater-embedded mechanically-controllable break junctions. Au microelectrodes were prepared on a polyimide-coated phosphor-bronze substrate by photolithography and radio-frequency (RF) magnetron sputtering followed by lift-off in $N,N$-
dimethylformamide (DMF) (i). After this, Al$_2$O$_3$ layers of thickness 25 nm were formed by electron-beam (EB) lithography and RF sputtering processes (ii). Then, a Au nanojunction was delineated between the two middle electrodes on the Al$_2$O$_3$ layer by EB lithography, and Au/Cr of thickness 100 nm/2 nm was deposited using the RF sputtering method (iii). After lift-off in DMF, a microheater was lithographically defined between the upper electrodes and 40 nm thick Pt layer was sputtered (iv). Finally, removing the resist in DMF, the substrate was exposed to isotropic reactive ion etching with oxygen etchant gas (v). As a result, a 2μm-long Au nanobridge was obtained with a microheater adjacent to it that can be used to impose temperature gradient for the thermopower measurements of atom-sized junctions.
Figure S2. Calibration of background voltage. Black plots are the average $\Delta V$, $\Delta V_{\text{ave}}$, obtained with no temperature gradient imposed ($V_h = 0$ V). The lateral axis is the conductance $G$ of Au atom-sized junctions. This background voltage was subtracted from $\Delta V_{\text{ave}}$ measured at $V_h > 0$ V.
Figure S3. Calculation of thermoelectric voltage. In the experiments, the potential drop $V_m$ at the 10 kΩ protective resistor was recorded under $V_b = 0$ V. The thermoelectric voltage at Au atom-sized contacts $\Delta V$ was deduced from $V_m$ after background subtraction by considering the voltage division at the contact of conductance $G$ as follows: $\Delta V = V_m(1 + 10^{-4}/G)$. 
Figure S4. Repeated measurements of the conductance $G$ (top) and the thermoelectric voltage $\Delta V$ (bottom) of Au atom-sized junctions. We recorded $\Delta V$ when $G$ becomes lower than $8 \, G_0$ during mechanical stretching of Au nanocontacts via substrate bending using the self-breaking technique. $G$ decreased in a stepwise manner reflecting discrete nature of the contact deformations. $\Delta V$ becomes zero when a contact breaks and $G$ drops to zero. After contact breakdown, we fused it up to $20 \, G_0$ by slowly releasing the bending force. Gray regions indicate the contact formation processes. Green and blue dashed lines shows $G = 1 \, G_0$ and $\Delta V = 0 \, V$ levels, respectively. Although fluctuating significantly, the thermoelectric voltage is positive at $G > 1 \, G_0$, whereas it occasionally takes negative values at lower $G$. It is also noticeable that $\Delta V$ rises sharply as $G$ decreases from $1 \, G_0$ to $0.8 \, G_0$ as denoted by the blue arrows.
Figure S5. Raw data of thermoelectric voltage. Black and red plots are the data and $\Delta V_{\text{ave}}$ of Au atom-sized junctions at $V_h = 2.0$ V, respectively. While fluctuates significantly, peak-like features in $\Delta V_{\text{ave}}$ are also observable in the raw data.
Figure S6. Calibration of a Pt microheater. \textbf{a,} False-colored scanning electron microscopy image of a microheater-embedded MCBJ. The substrate temperature $T_0$ was altered in a range from 293 K to 400 K using an electrical
heater and a thermometer attached to the sample holder. Meanwhile, the resistance of the Pt heater $R$ was recorded under a small voltage $V_h = 0.1$ V using a picoammeter-source unit. As seen in the figure, $R$ tends to increase almost linearly at a rate $R = 3.7 T_0 + 5686 \, \Omega$, which is similar to the previous reports. After obtaining the calibration curve, $R - V_h$ characteristics were measured from which we deduced the temperature at the heater under an elevated $V_h$. The $T_h - V_h$ characteristics reveal monotonic increase of the heater temperature with $V_h^2$ suggestive of Joule heat dissipation at the current-carrying Pt coil.
Figure S7. Average thermoelectric voltage $\Delta V_{\text{ave}}$ in a low-$V_h$ regime below 2 V. Conductance $G$ is not corrected with $R_s = 800$ $\Omega$ (see main text). The oscillation behavior appears when the heater voltage is increased to 0.6 V and becomes more evident at higher $V_h$. 
Figure S8. Average thermoelectric voltage $\Delta V_{\text{ave}}$ above $V_h = 2$ V showing conductance-dependent oscillation at below $3 \, G_0$. Conductance $G$ is not corrected with $R_s = 800 \, \Omega$ (see main text).
Figure S9. Two dimensional conductance histogram constructed with 50 traces below 1 $G_0$ at $V_h = 2.4$ V. It shows that the single-atom contact is stretched for not more than 20 pm and breaks down via thermal fluctuation under the very low stretching speed, 0.6 pm/s.
Figure S10. Influence of Au leads on the thermoelectric voltage measurements. The voltage drop at III can be described as $(V_c - V_0) = S_c(T_c - T_0)$, where $S_c$ is the thermopower of the atom-sized junction. In addition to this, temperature gradient along the long Au leads creates a thermovoltage that contributes to the measured $\Delta V = V_2 - V_0$. This can be defined as $(V_1 - V_c) = S_{Au}(T_1 - T_c)$ and $(V_2 - V_1) = S_{Au}(T_2 - T_1)$ in the region II and I, respectively, where $S_{Au} = 1.94 \mu V/K$ is the thermopower of bulk Au. Here, $T_2$ is the temperature at the end of the millimeter-long Au lead, which should be not so different than $T_0$. As a result, $V_2 - V_0 = \Delta V \sim (S_{Au} - S_c)(T_c - T_0) = (S_{Au} - S_c) \Delta T$. 
2. Supplementary references


