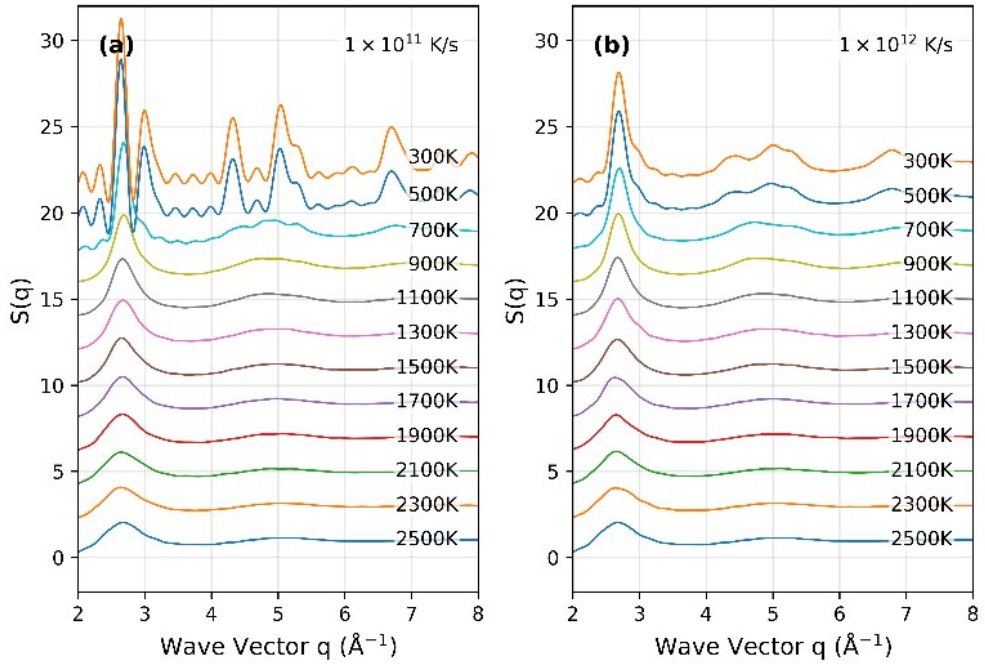

Figure S1.

Comparison of the evolution of the static structure factor $S(q)$ of gold atoms with temperature. The curves are vertically offset for clarity to illustrate the changes in $S(q)$ at different temperatures during cooling from 2500 K to 300 K. The corresponding temperature values are indicated at the right end of each curve. (a) Cooling rate of 1×10^{11}K/s; (b) Cooling rate of 1×10^{12}K/s.

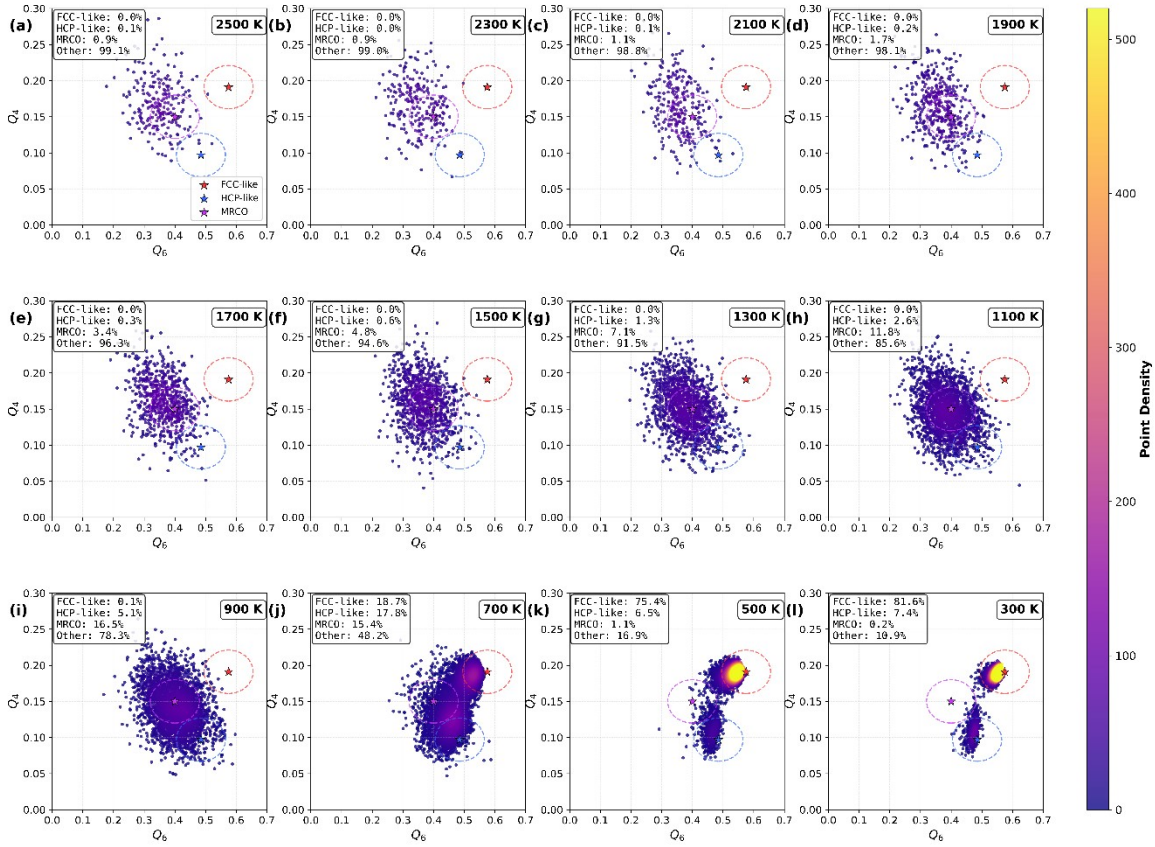

Figure S2a.

Q_6–Q_4 distribution maps showing the evolution of local structural types in the system during cooling from 2500 K to 300 K at a cooling rate of 1×10^{11} K/s. The scatter plots are constructed based on the joint distribution of atomic local bond-orientational parameters Q_6 and Q_4, with color indicating point density calculated by Gaussian kernel density estimation, with higher density representing more atoms with similar local structures. Asterisks mark the centers of standard crystalline structures: FCC-like ($Q_6 = 0.575$, $Q_4 = 0.191$), HCP-like ($Q_6 = 0.485$, $Q_4 = 0.097$), and the empirically defined MRCO region (around $Q_6 \approx 0.4$, $Q_4 \approx 0.15$). Dashed ellipses indicate the identification regions for each structure (tolerances of $Q_6 \pm 0.08$ and $Q_4 \pm 0.03$).

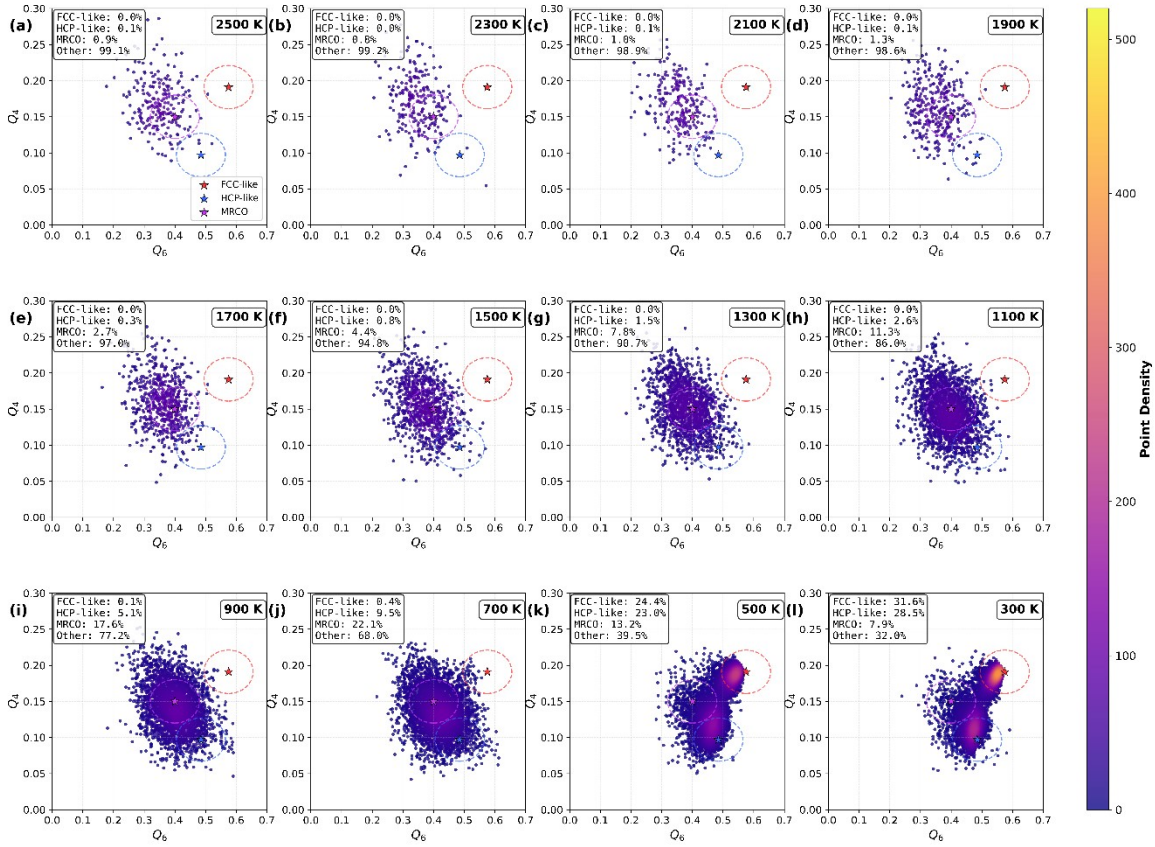

Figure S2b.

Q_6–Q_4 distribution maps showing the evolution of local structural types in the system during cooling from 2500 K to 300 K at a cooling rate of 1×10^{12} K/s. The scatter plots are constructed based on the joint distribution of atomic local bond-orientational parameters Q_6 and Q_4, with color indicating point density calculated by Gaussian kernel density estimation, with higher density representing more atoms with similar local structures. Asterisks mark the centers of standard crystalline structures: FCC-like ($Q_6 = 0.575$, $Q_4 = 0.191$), HCP-like ($Q_6 = 0.485$, $Q_4 = 0.097$), and the empirically defined MRCO region (around $Q_6 \approx 0.4$, $Q_4 \approx 0.15$). Dashed ellipses indicate the identification regions for each structure (tolerances of $Q_6 \pm 0.08$ and $Q_4 \pm 0.03$).

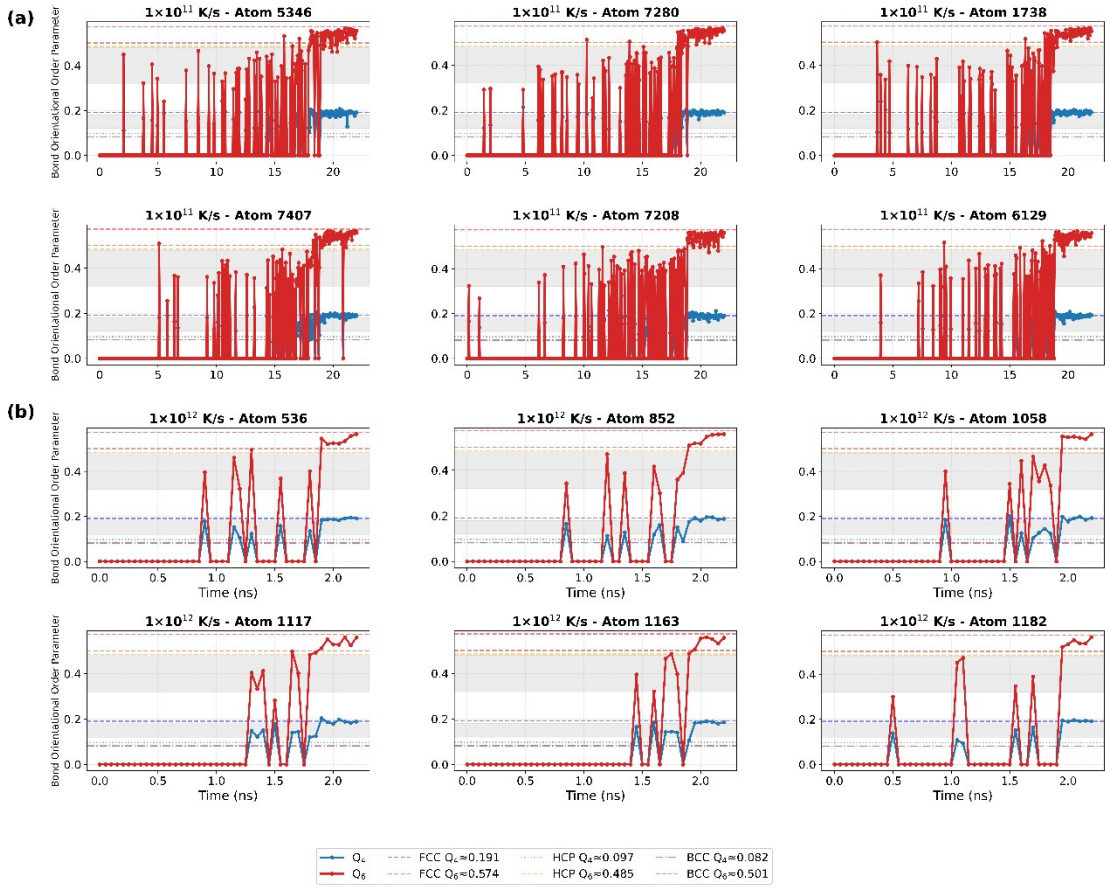

Figure S3

Evolution of bond order parameters of selected atoms under different cooling rates. (a) At a cooling rate of 1×10^{11} K/s, the Q_4 and Q_6 parameters of other representative atoms (ID: 5346, 7280, 1738, 7407, 7208, 6129) evolve over time during cooling from 2500 K to 300 K. (b) At a cooling rate of 1×10^{12} K/s, the corresponding evolution of other representative atoms (ID: 536, 852, 1058, 1117, 1163, 1182) during cooling from 2500 K to 300 K. The shaded regions (0.32–0.48 and 0.12–0.18) indicate the medium-range crystalline order (MRCO) region. Dashed and dash-dotted lines represent the reference values for FCC ($Q_4 \approx 0.191$, $Q_6 \approx 0.574$), HCP ($Q_4 \approx 0.097$, $Q_6 \approx 0.485$), and BCC ($Q_4 \approx 0.082$, $Q_6 \approx 0.501$) structures, respectively.

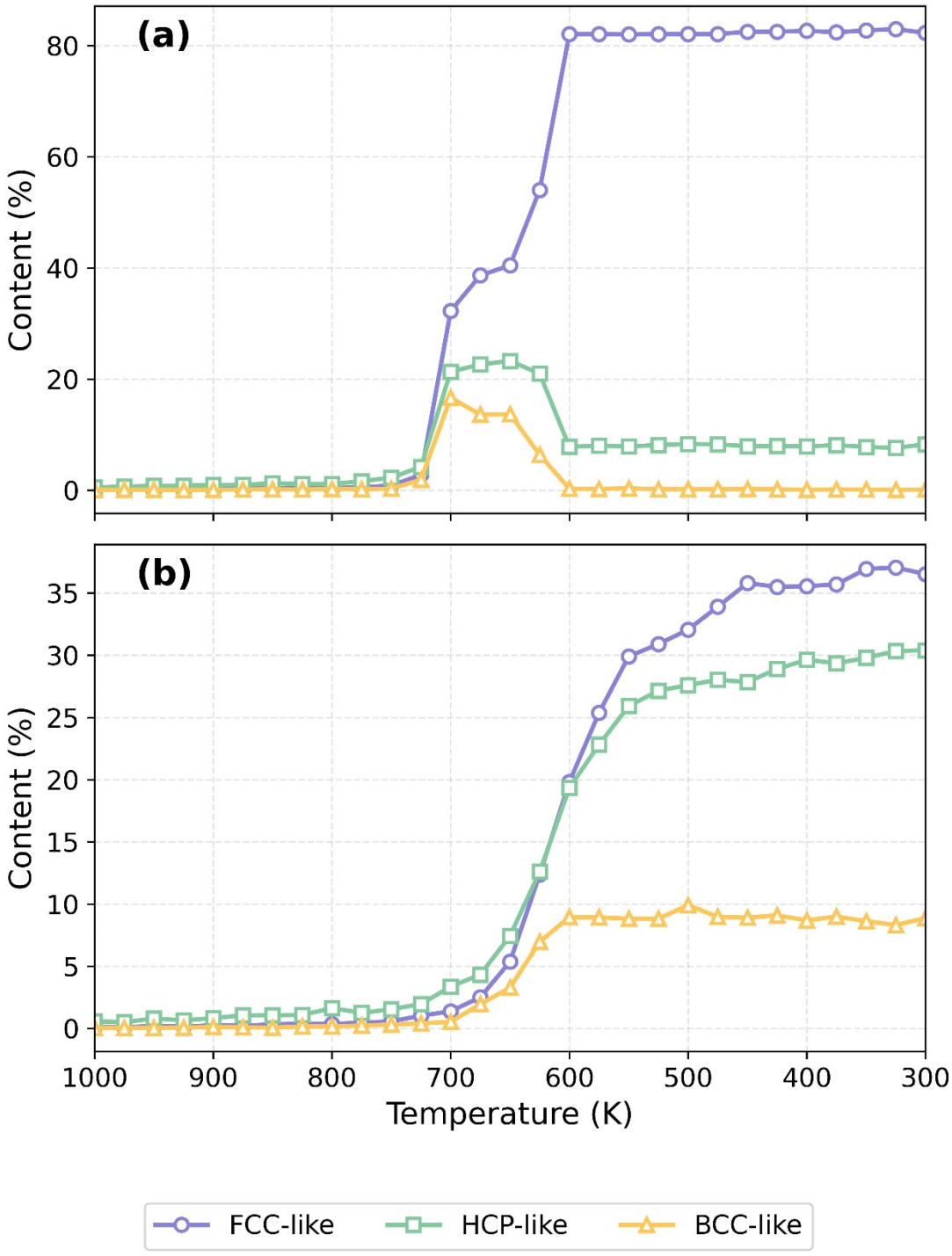

Figure S4.

Comparative plot of the temperature-dependent content of gold atom local structures based on PTM analysis (RMSD = 0.15), corresponding to cooling rates of (a) 1×10^{11} K/s and (b) 1×10^{12} K/s. Each curve represents the percentage of different structural types (FCC-like, HCP-like, BCC-like) as temperature decreases from 1000 K to 300 K. The figure reveals the competitive evolution of different local structures under different cooling rates.

Table S1. MRCO Residence Time Statistics at Cooling Rate of 10^{11} K/s

Atom ID	MRCO Events	Total Residence Time (ns)	Average Residence Time (ns)	Fraction of Total Time (%)
7476	29	1.7	0.059	7.7
697	34	2.35	0.069	10.7
4825	41	2.75	0.067	12.5
7362	36	2.35	0.065	10.7
5346	38	2.35	0.062	10.7
7280	32	2.2	0.069	10
1738	35	2.25	0.064	10.2
7407	32	2.9	0.091	13.2
7208	45	3.2	0.071	14.5
6129	34	2.4	0.071	10.9
Mean ± SD	35.6 ± 4.8	2.45 ± 0.43	0.069 ± 0.009	11.1 ± 1.9

Note: Total cooling time = 22 ns. Cooling rate: 10^{11} K/s.

Table S2. MRCO Residence Time Statistics at Cooling Rate of 10^{12} K/s

Atom ID	MRCO Events	Total Residence Time (ns)	Average Residence Time (ns)	Fraction of Total Time (%)
368	2	0.1	0.05	4.5
403	6	0.35	0.058	15.9
441	6	0.45	0.075	20.5
530	3	0.2	0.067	9.1
536	5	0.3	0.06	13.6
852	5	0.3	0.06	13.6
1058	4	0.35	0.088	15.9
1117	3	0.25	0.083	11.4
1163	3	0.3	0.1	13.6
1182	4	0.25	0.063	11.4
Mean ± SD	4.1 ± 1.4	0.285 ± 0.10	0.070 ± 0.016	13.0 ± 4.4

Note: Total cooling time = 2.2 ns. Cooling rate: 10^{12} K/s.