Supporting Information

Origin of p-type conduction in amorphous CuI: a first-principles investigation

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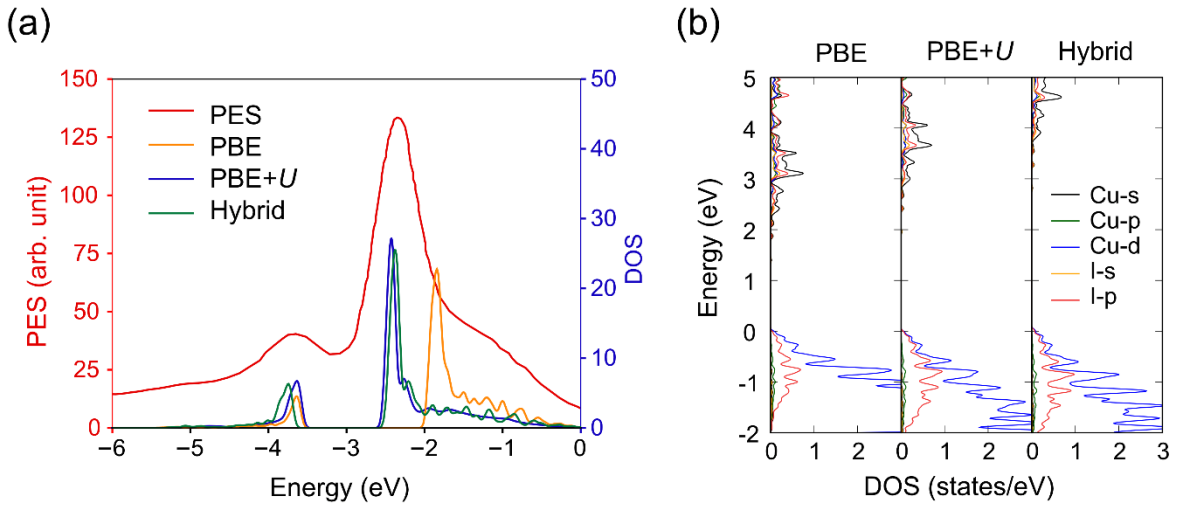

Figure S1. (a) A comparison for Cu-3d level of γ-CuI between experiment and theory. (b) Partial density of states of γ-CuI calculated using PBE, PBE+*U*, and Hybrid functionals.

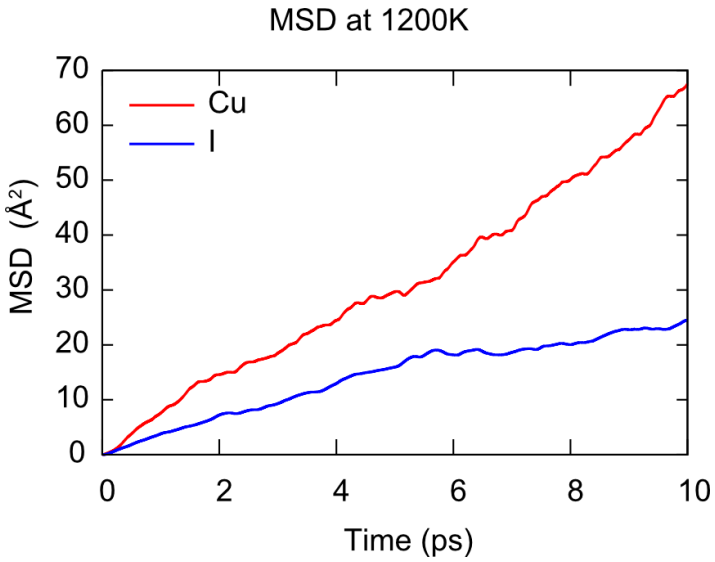

Figure S2. The ion-averaged mean square displacement of Cu and I ions.

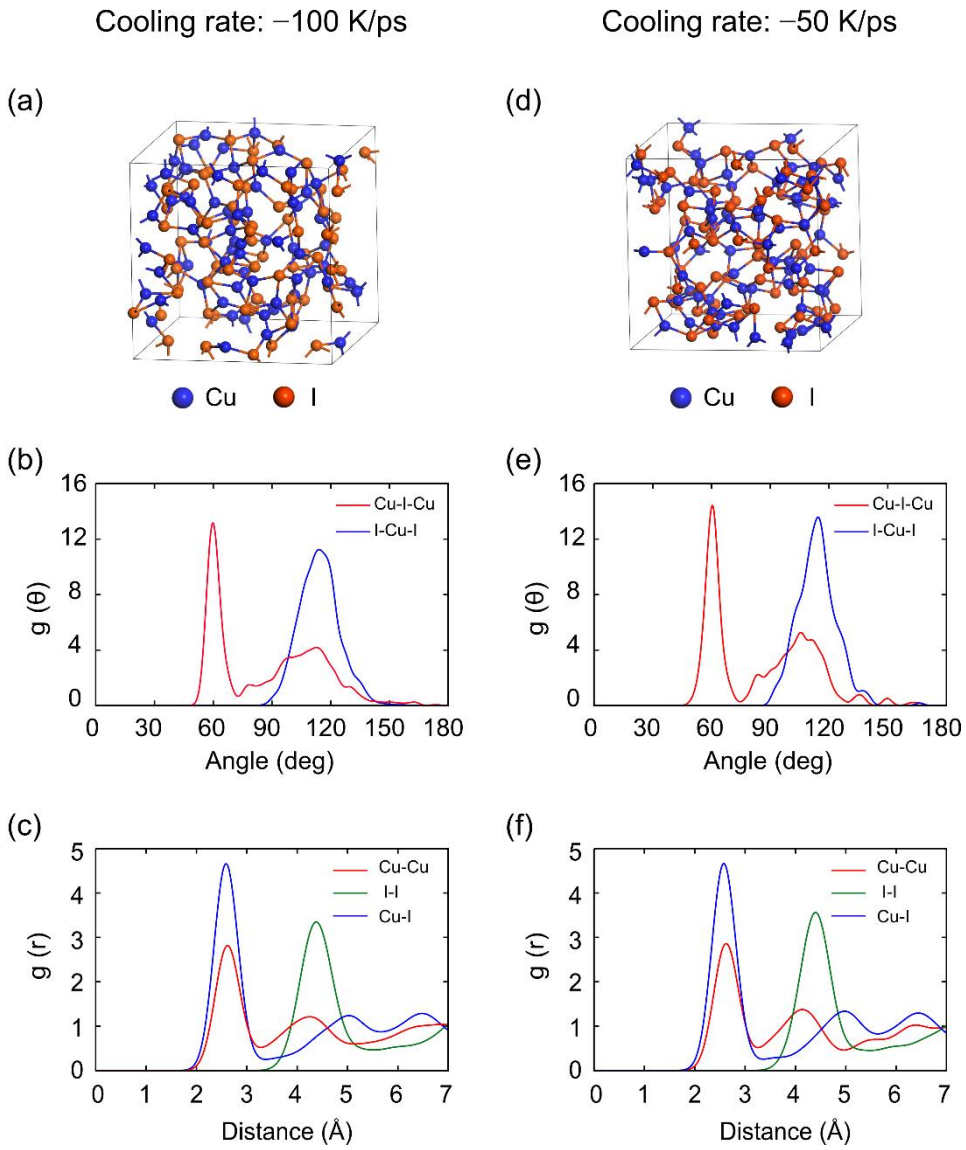

Figure S3. Impact of quenching rate on the amorphous structures. The atomic structure, ADFs and RDFs for different cooling rates. (a)-(c): −100 K/ps and (d)-(f): −50 K/ps.

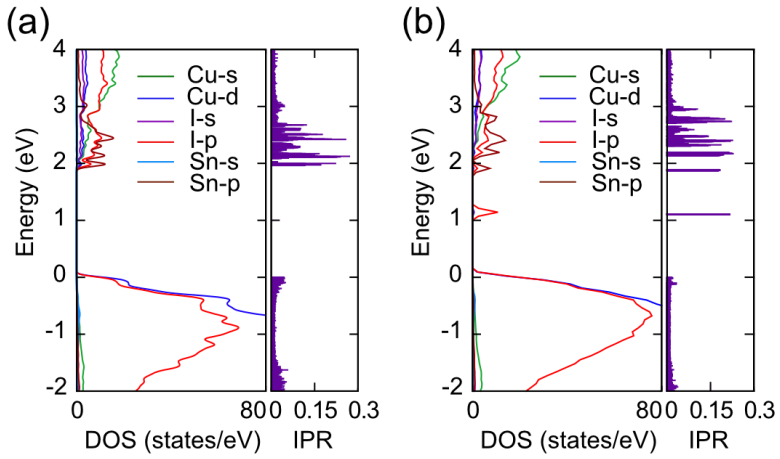

Figure S4. Difference in electronic structures depending on Sn charge states. PDOSs and IPR

for (a) *a*-CuI:Sn^{2+} and (b) *a*-CuI:Sn^{4+}. We model amorphous structures including Sn^{2+} and Sn^{4+} by adding two I atoms per one Sn atom (N_{Cu}:N_I:N_{Sn} = 72:4:80) and four I atoms per one Sn atom (N_{Cu}:N_I:N_{Sn} = 72:4:88), respectively. We find that Bader charges of three of four Sn ions in the latter are about 2.97 that are higher than 2.54 for Sn ions in the former, consistent with our expectation. On the other hand, one Sn ion in the supercell including Sn^{4+} displays Bader charge of 2.52 which is less than the others, indicating it loses smaller electrons. This happens because covalent bonds are formed among some I ions, producing a mid-gap state.

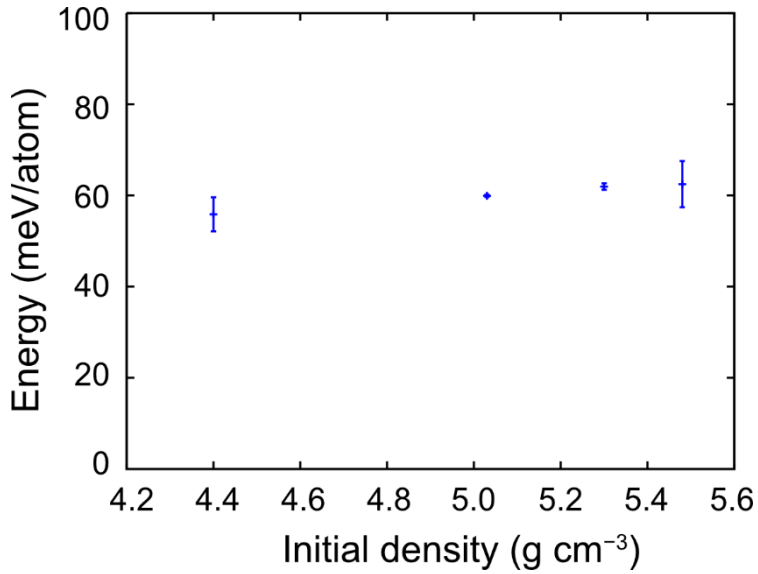

Figure S5. Initial density vs. their averaged energy of initial density from 4.44 g cm^{-3} to 5.48 g cm^{-3}.

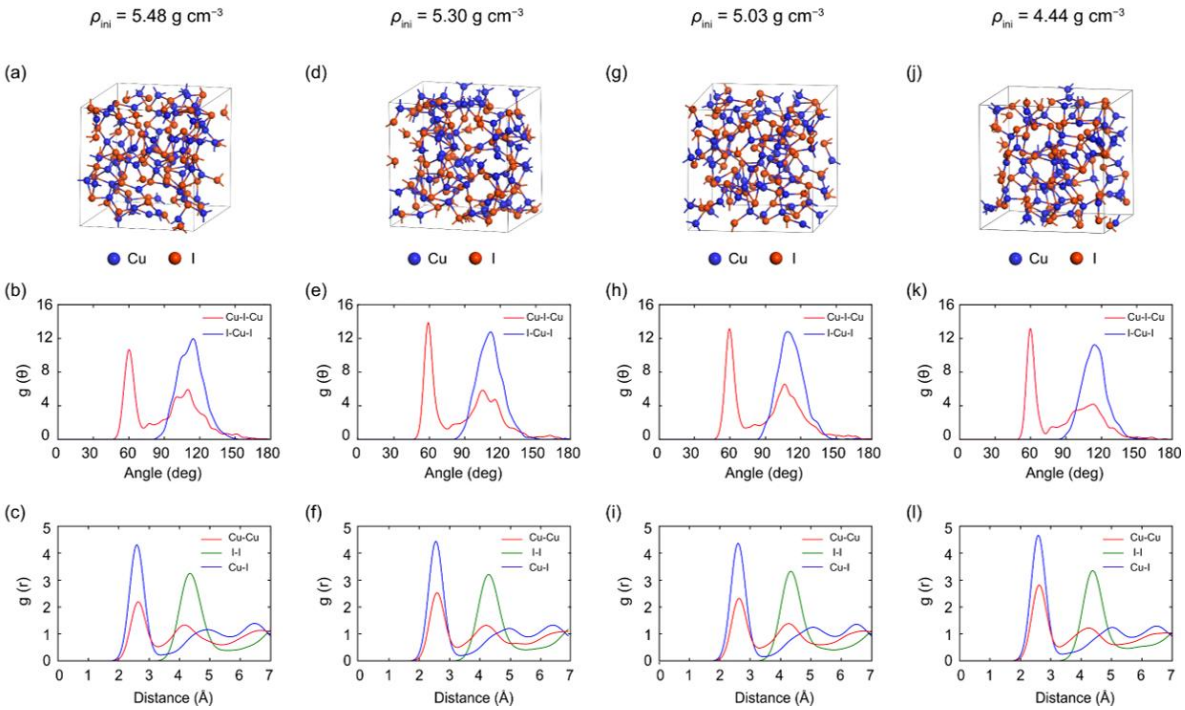

Figure S6. A comparison of various initial density. The atomic structure, ADFs and RDFs for each initial mass density. (a)-(c): 5.48 g cm^{-3}, (d)-(f): 5.30 g cm^{-3}, (g)-(i): 5.03 g cm^{-3} and (j)-

(l): 4.44 g cm^{-3}.

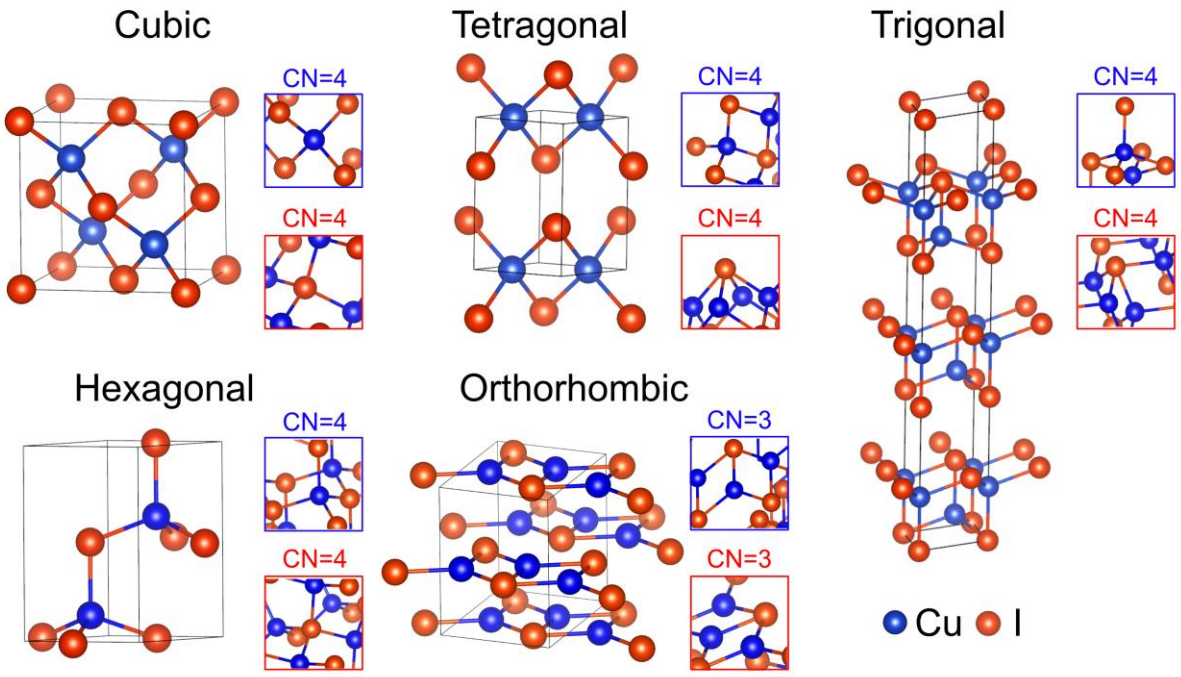

Figure S7. The atomic structures and their local structures of CuI polymorphs.

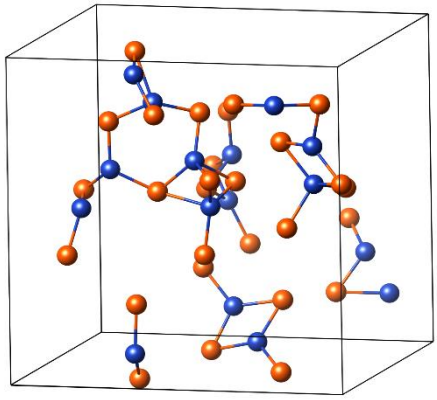

Figure S8. Cu-I bonds for Cu ions with Cu neighbors within 2.55 Å.

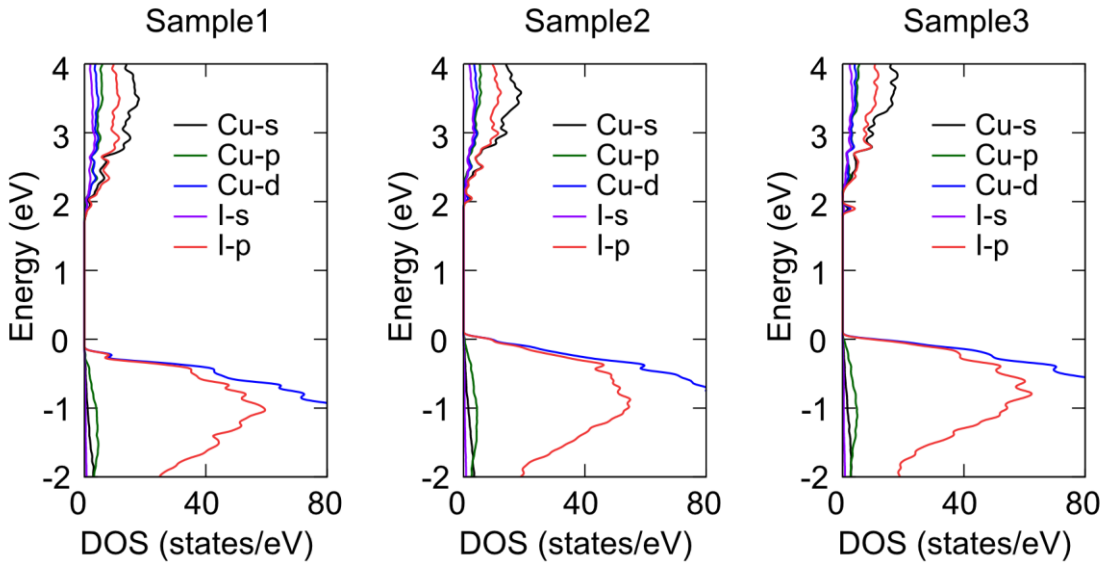

Figure S9. Density of states for three *a*-CuI structures.

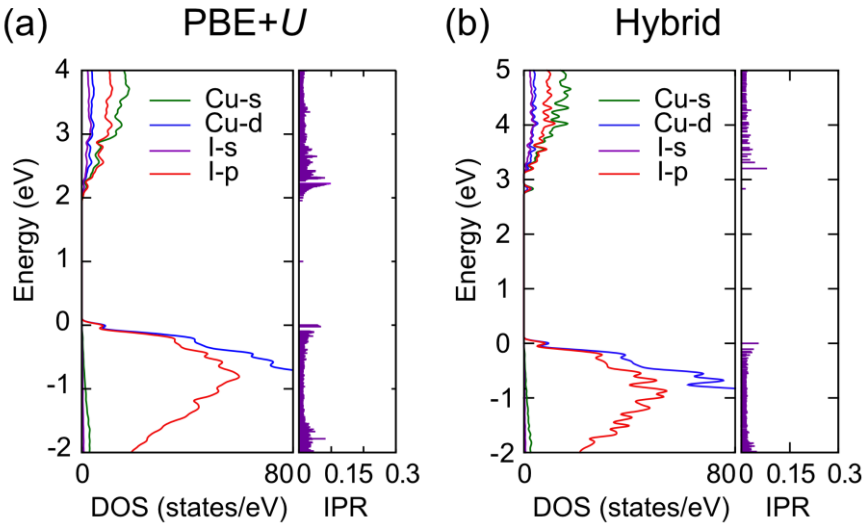

Figure S10. DOSs and IPR for *a*-CuI in (a) PBE+*U* and (b) hybrid scheme.

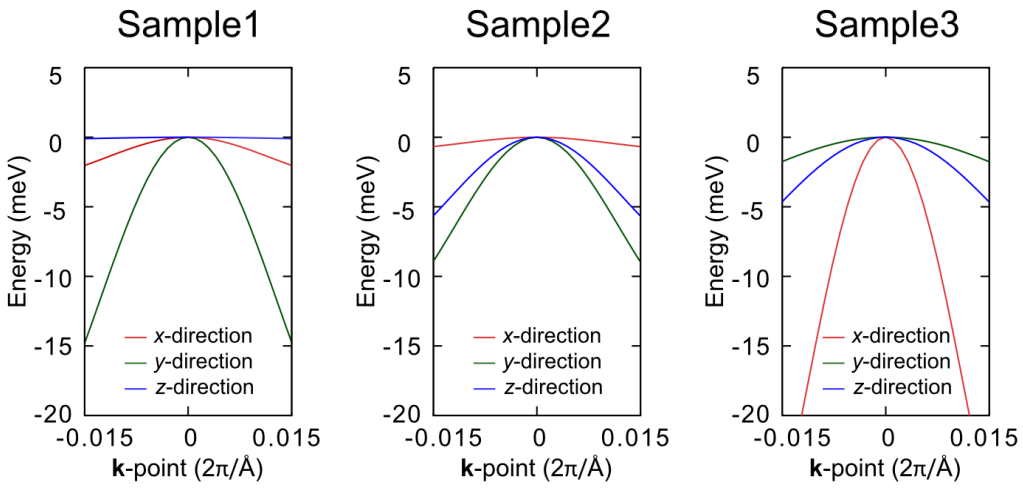

Figure S11. Band dispersions for three *a*-CuI structures.