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Supporting Information

Effects of local geometry distortion at the Al/Al_2Cu interfaces on solute segregation

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The supercells to simulate small lattice mismatch are pretty large (2.29%: a=28.32 Å and containing 203 atoms; 0.25%: a=39.86 Å and containing 287 atoms). Moreover, for each doping atom (Si, Mg and Zn), there are 64 and 88 non-equivalent substitution sites for the lattice mismatch of 2.29% and 0.25%, respectively. In other words, we need to perform 64×3=192 and 88×3=264 full geometry relaxations in large supercells for the lattice mismatch of 2.29% and 0.25%, respectively. It is time consuming even using the OpenMX-NCPP-LCAO methodology. Nevertheless, we do the following tests: Using the lattice mismatch of 2.29%, we calculate the segregation energies of Si at Al and Al_2Cu sides in the Al/Al_2Cu semicoherent interface region, and find that the lowest segregation energies of Si at the Al and Al_2Cu side are -0.22 and -0.41 eV, respectively. They are lower than the corresponding segregation energies of Si in the coherent interface region (-0.10 eV and -0.31 eV). This means that the Si concentration in the semicoherent region is higher than that in the coherent interface region. The calculated results are consistent with results either from the

experimental measurements or using a larger lattice mismatch (4.52% with a smaller supercell). Thus, using the smaller supercell of 4.52% lattice mismatch is enough to efficiently address the Al/Al_2Cu problems and gives rise to the same conclusions as using large supercell of low lattice mismatch.

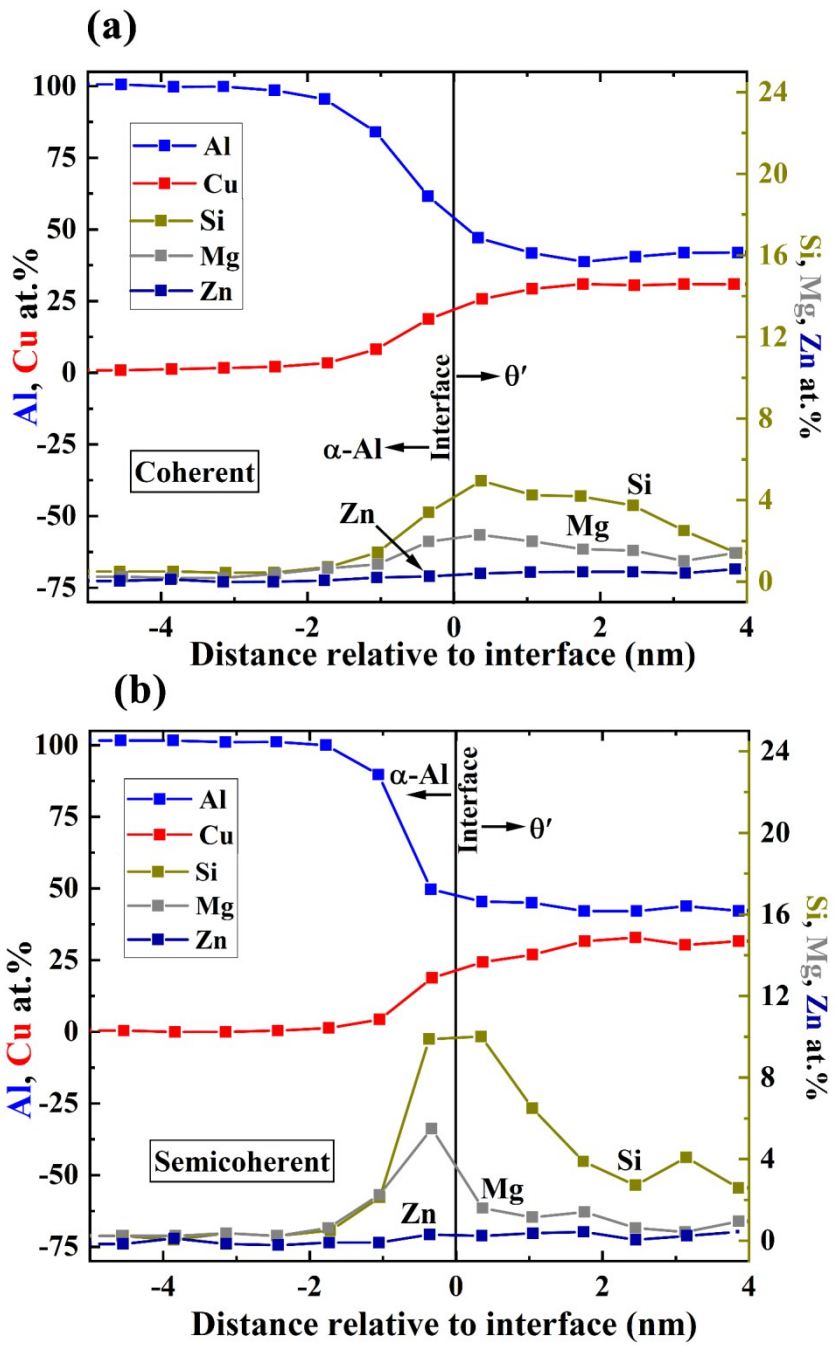

Fig. S1 Concentrations of Al, Cu, Mg, Si, and Zn as a function of distance from the coherent (a) and semicoherent (b) α-Al/θ′ matrix-precipitate interfaces after aging at 463 K for 8 h. Al and Cu levels are identified on the left ordinates; Si, Mg, and Zn levels appear on the right. The location of each interface is depicted with a vertical line. (The data are taken from ref. [1])

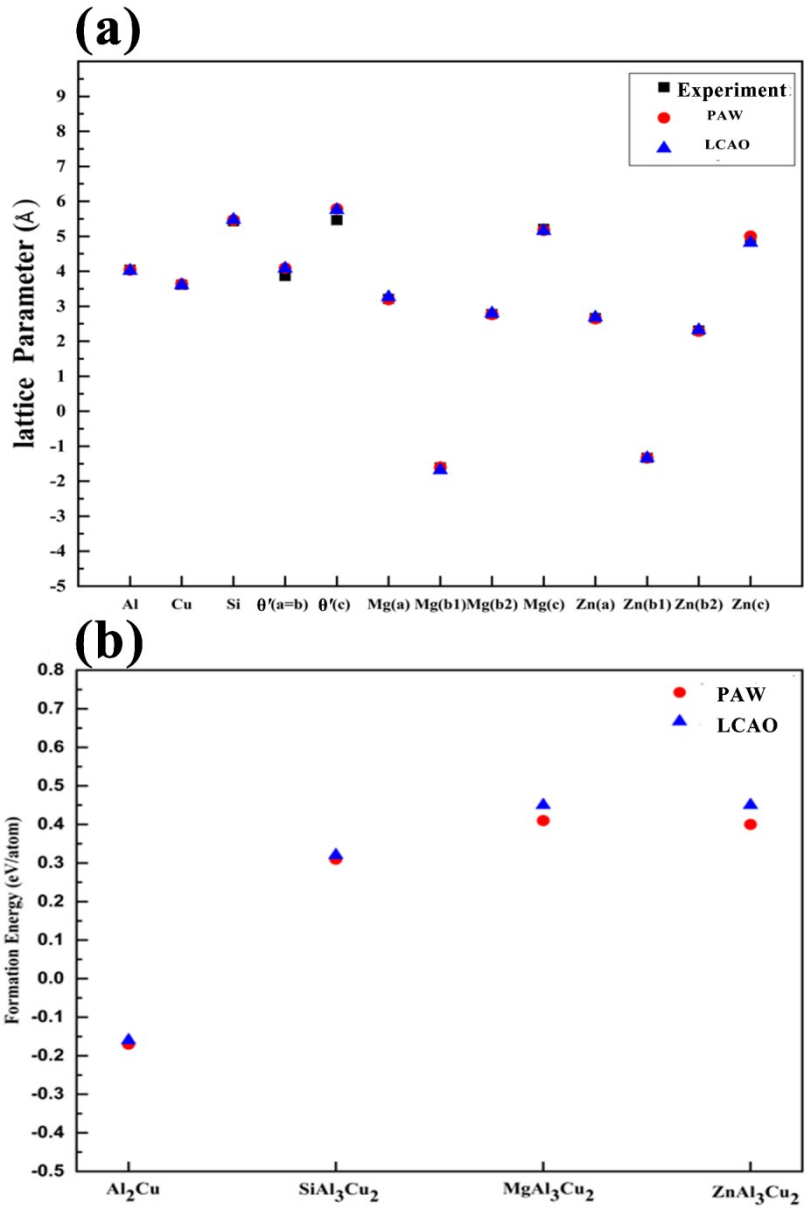

Fig. S2 Calculated lattice parameters of bulks [Al, Cu, Al_2Cu (a=b), Si, Mg (a, b_1, b_2, c), Zn (a, b_1, b_2, c)] (a) and formation energy for XAl_3Cu_2 (X=Si, Mg Zn) using LCAO (blue triangles) and PAW (red circles) (b).

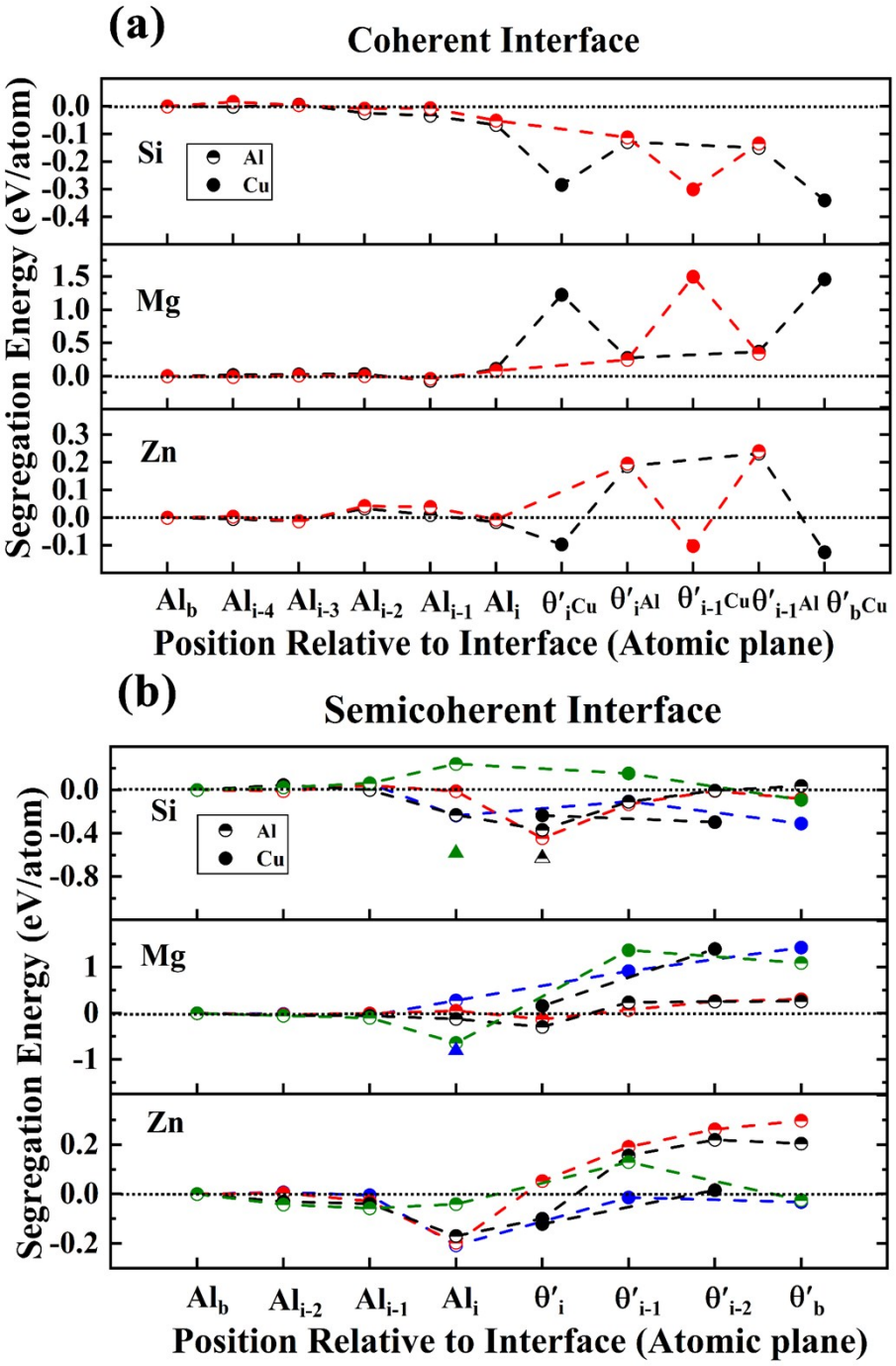

Fig. S3 Calculated segregation energies of solutes for Si, Mg and Zn with the position relative to Al/Al_2Cu coherent (a) and semicoherent (b) interfaces using the PAW method. The semi-filled and filled circles represent substitution at Al and Cu sites, respectively. The green (blue, semi-filled black) triangle in (b) represent the segregation energies of Si (Mg) in the Al_i and θ′ i layers obtained by further relaxing the LCAO output lowest energy structures using PAW.

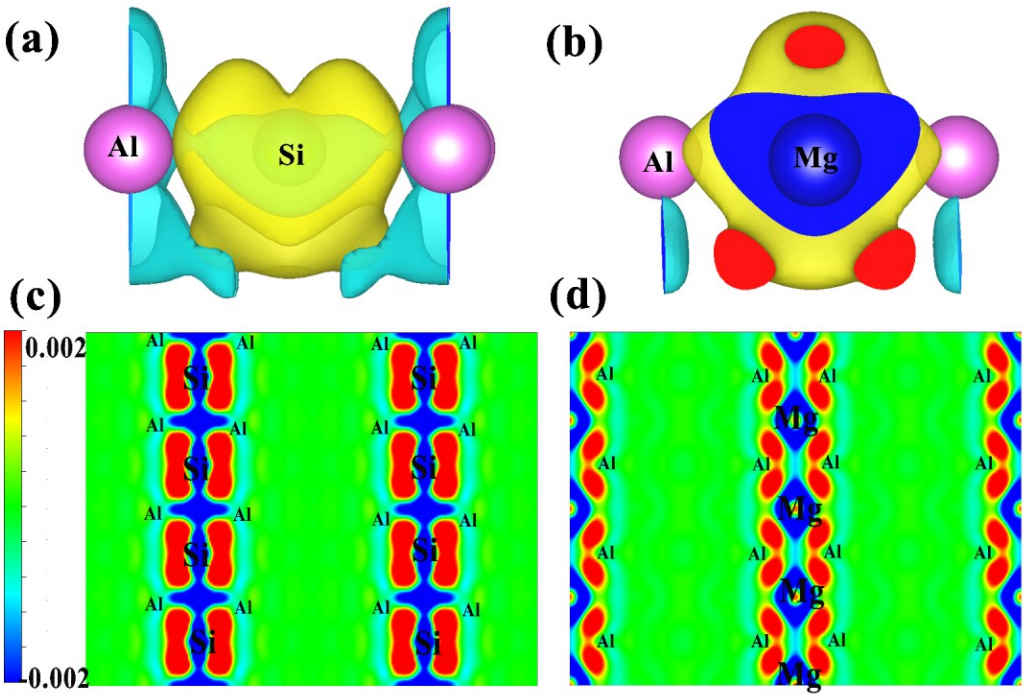

Fig. S4 3D and contour on the (001) plane of charge density difference of Si (a, c) and Mg (b, d) at the Al_i layer (Fig. 2b) in the Al/Al_2Cu semicoherent interface using PAW. (The isosurface value is 0.002 eV/bohr^3). Yellow and green regions denote charge accumulation and depletion, respectively.

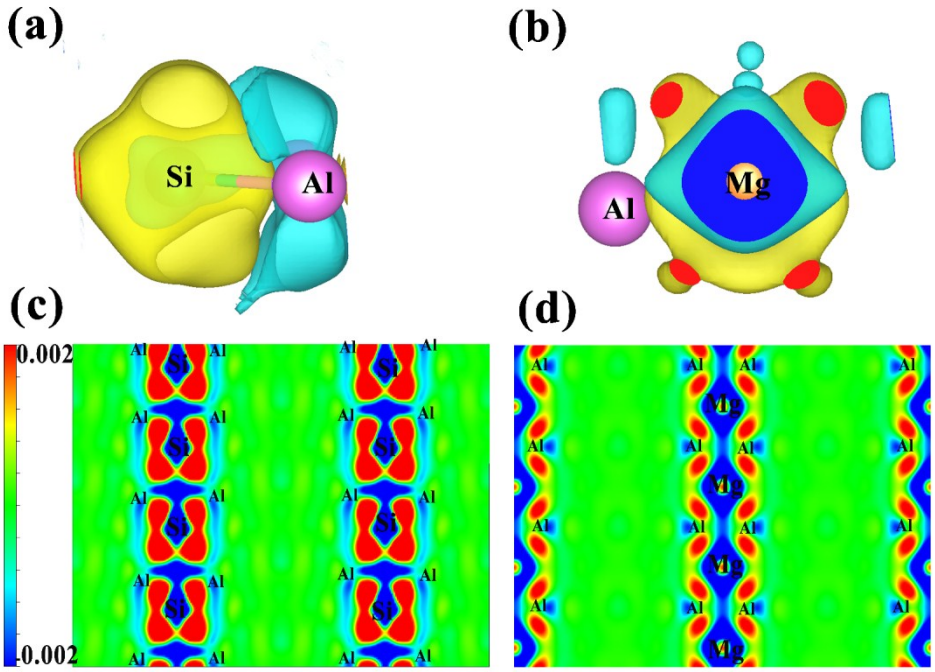

Fig. S5 3D and contour on the (001) plane of charge density difference of Si (a, c) and Mg (b, d) at Al_i layer (Fig. 2b) in the Al/Al_2Cu semicoherent interface by further relaxing the LCAO output lowest energy structures using PAW. (The isosurface value is 0.002 eV/bohr^3). Yellow and green regions denote charge accumulation and depletion, respectively.

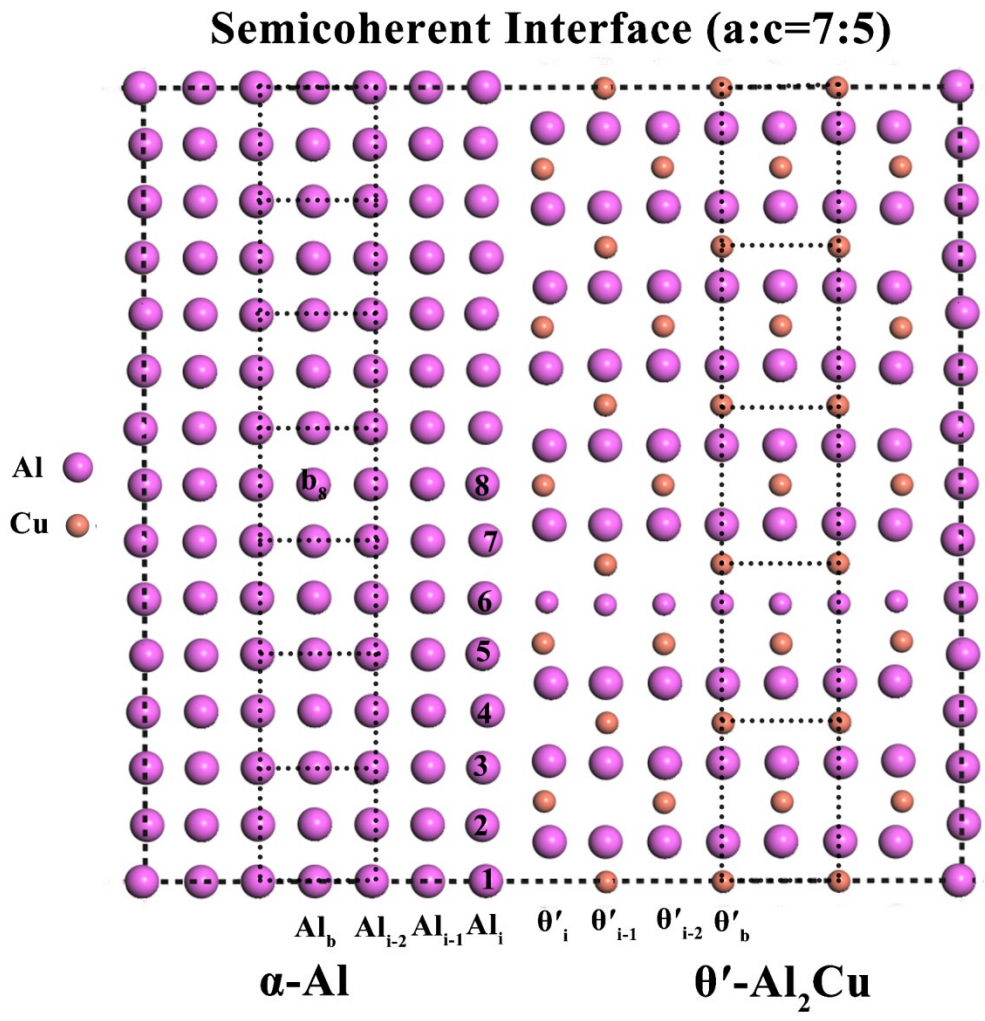

Fig. S6 Supercell slab models to simulate the segregation energies of Si (Mg) in the $(010)_{Al}$|| $(010)_{\theta'}$ semicoherent interface with a mismatch of 2.29% utilizing LCAO. Large and small spheres represent Al and Cu atoms. Dashed lines mean the $7a_{Al}=5c_{\theta'}$ relationships of the semicoherent interface. The numbers from 1 to 8 represent the substitutional sites for solutes in the Al_i layer.

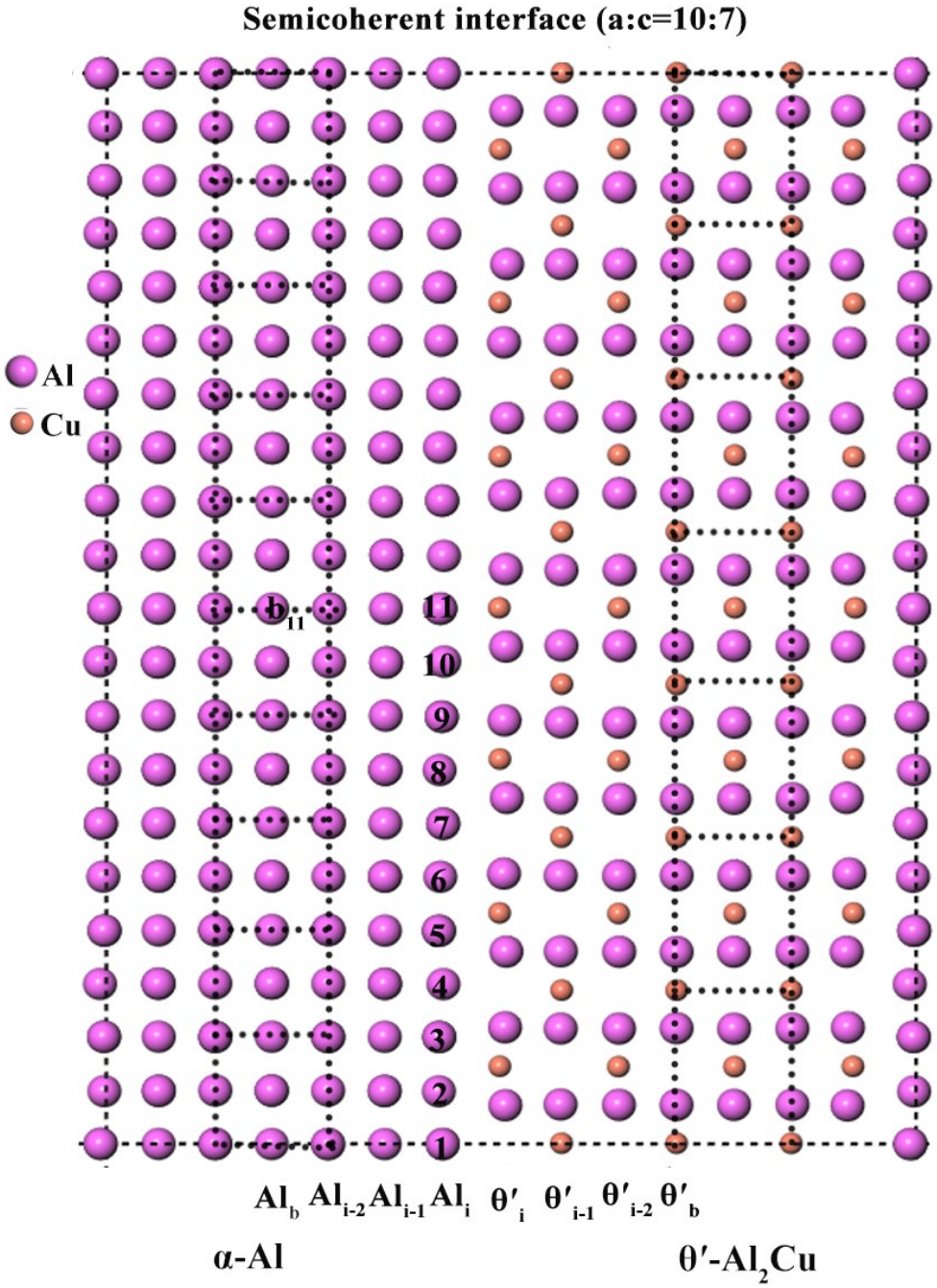

Fig. S7 Supercell slab models to simulate the segregation energies of Si (Mg) in the $(010)_{Al}$‖ $(010)_{\theta'}$ semicoherent interface with a mismatch of 0.25% utilizing LCAO. Large and small spheres represent Al and Cu atoms. Dashed lines mean the $10a_{Al}=7c_{\theta'}$ relationships of the semicoherent interface. The numbers from 1 to 11 represent the substitutional sites for solutes in the Al_i layer.

Table S1 Theoretically (LCAO and PAW) calculated lattice parameters (a, b, c) for Al/Al_2Cu coherent and semicoherent interfaces.

	Coherent interface			Semicoherent interface		
	a (Å)	b (Å)	c (Å)	a (Å)	b (Å)	c (Å)
PAW	4.04	4.04	35.20	11.73	4.06	28.95
LCAO	4.03	4.03	34.80	11.75	4.04	28.79

Table S2 Interface spacing (Å) between two phases (Al and Al_2Cu) in the semicoherent interface with and without Si (Mg) solute using LCAO and PAW.

	Solute		Clean interface
Structure	Si (Al_i green dot)	Mg (Al_i blue dot)	
LCAO (Å)	2.22	2.19	2.17
PAW (Å)	2.32	2.23	2.21

Table S3 Calculated segregation energy of solutes (Si, Mg) at the different sites (Fig. S6) in the semicoherent interface with the misfit of 2.29% using LCAO. **Al_2 (Al_1)** and **Al_3 (Al_4)** are low1 and low2 structure for solute Si (Mg) in Table 1, respectively.

	Segregation energy at Al_i substitutional sites (eV/atom)							
Solute	Al_1	Al_2	Al_3	Al_4	Al_5	Al_6	Al_7	Al_8
Si	-0.17	**-0.20**	**-0.22**	-0.20	-0.19	-0.12	-0.13	-0.12
Mg	**-0.35**	0.10	0.11	**-0.30**	0.08	-0.12	-0.27	0.20

Table S4 Calculated segregation energy of solutes (Si, Mg) at the different sites (Fig. S7) in the semicoherent interface with the misfit of 0.25%. **Al_7 (Al_6)** and **Al_{10} (Al_9)** are low1 and low2 structure for solute Si (Mg) in Table 1, respectively.

	Segregation energy at Al_i substitutional sites (eV/atom)					
Solute	Al_1	Al_2	Al_3	Al_4	Al_5	Al_6
Si	-0.22	-0.20	-0.21	-0.24	-0.18	-0.22
Mg	-0.33	0.13	0.13	-0.31	-0.03	**-0.45**
Solute	Al_7	Al_8	Al_9	Al_{10}	Al_{11}	
Si	**-0.28**	-0.19	-0.21	**-0.31**	-0.13	
Mg	-0.29	0.10	**-0.53**	-0.30	0.09	

1. A. Biswas, D. J. Siegel and D. N. Seidman, *Phys. Rev. Lett.*, 2010, **105**, 076102.