Supplemental Materials for

Revealing pressure-induced softening/weakening mechanism in representative covalent materials

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Fig. S1. Calculated phonon dispersion curves of (a) diamond, (b) c-BN, (c) Si and (d) Ge under zero and high pressures.

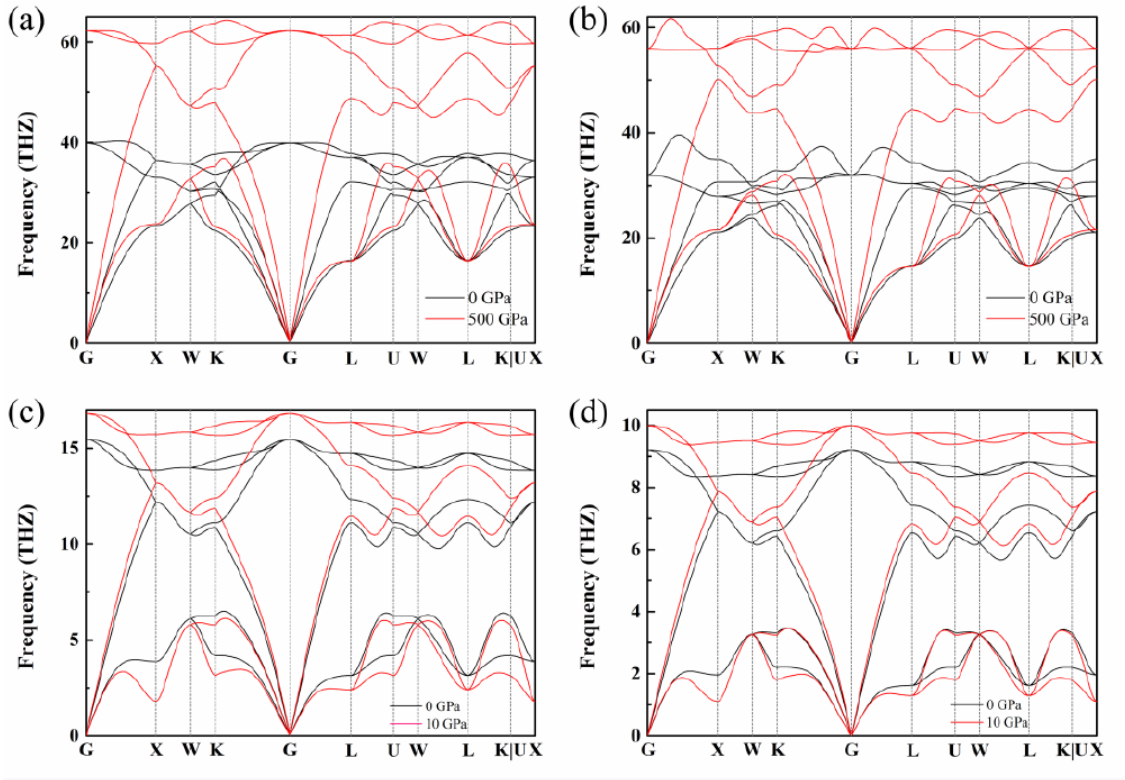

Fig. S2. Calculated isosurfaces of the VCDD of (a1)-(a9) diamond, (b1)-(b9) c-BN, (c1)-(c9) Si and (d1)-(d9) Ge under different strains along the (111)[11$\bar{2}$] shearing path under different pressures. The unit of VCDD is electrons/Bohr^3, and a same isosurface level of ±0.0305, ±0.0340, ±0.0375 electrons/Bohr^3 is used for diamond under 300, 400 and 500 GPa, ±0.022, ±0.033, ±0.036 for c-BN under 100, 400 and 500 GPa, ±0.008 for Si and ±0.005 for Ge under 0, 2, 10 GPa, respectively. Blue and yellow regions signify the states of charge depletion (negative) and accumulation (position), respectively.

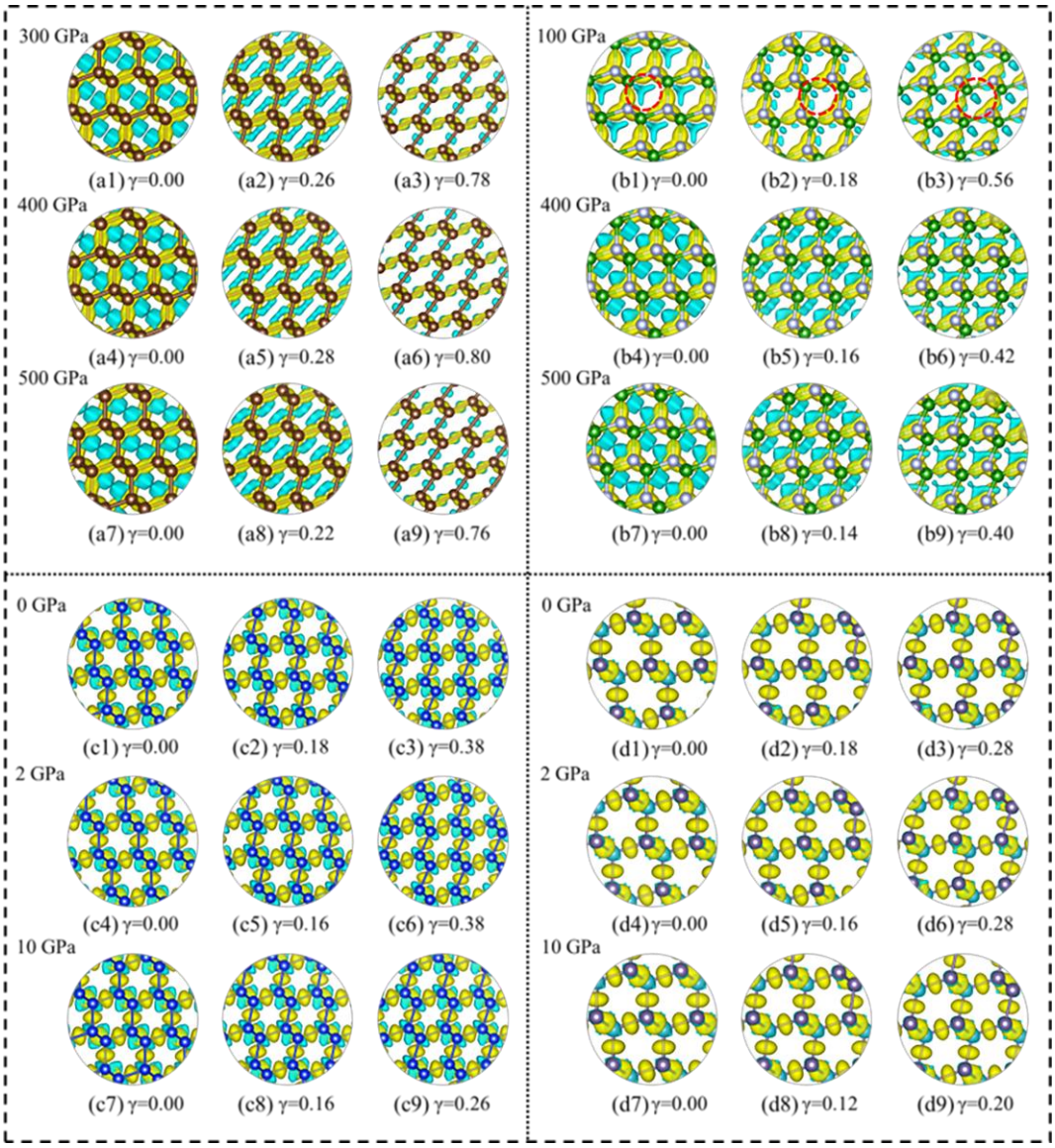

Fig. S3. Fat band structures and the corresponding DOS curves of diamond slipping with various displacements (γ=0.00, 0.45) along the shuffle-set plane at different pressures.

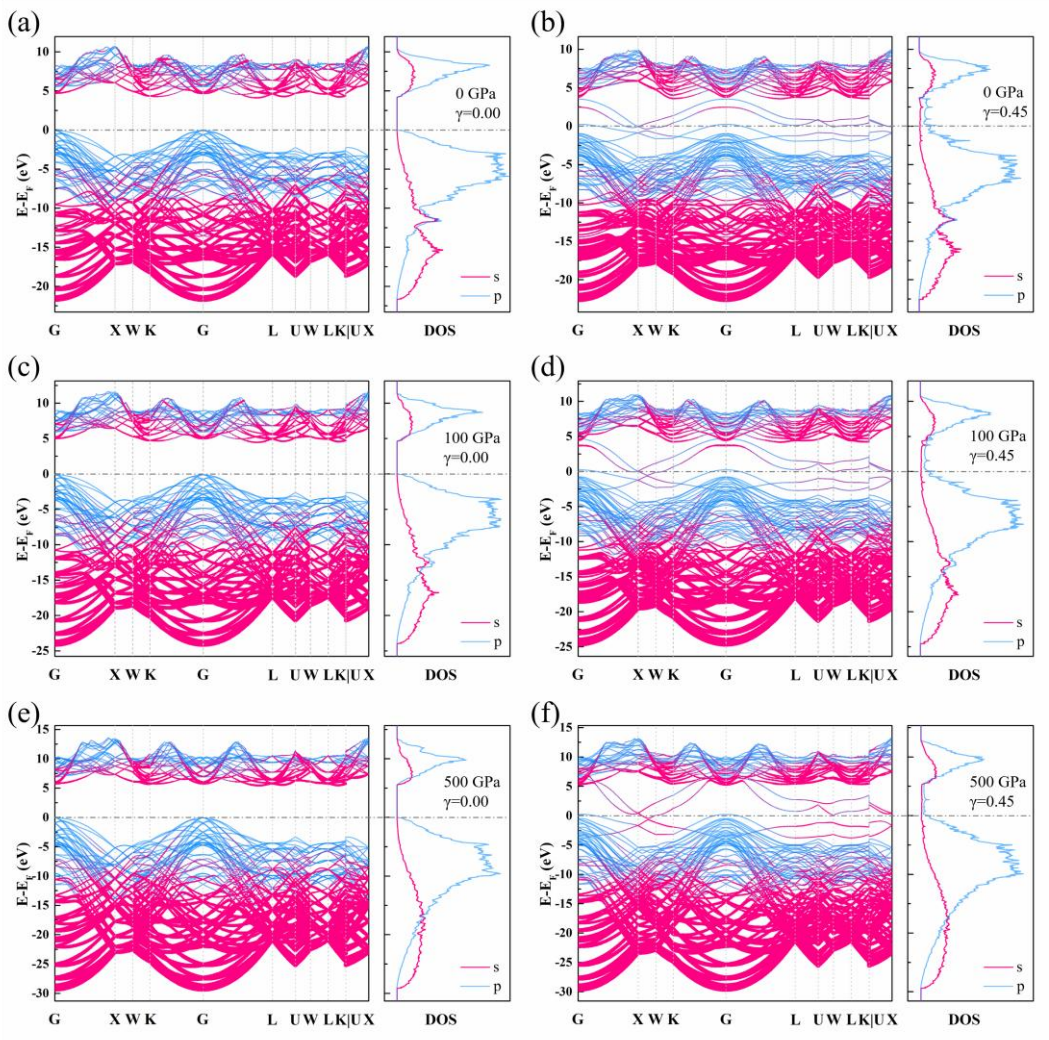

Fig. S4. The COHP curves of the bonds around the slip plane "S0" in (a) Si and (b) Ge under various displacements (u=0.00 and u=0.45) at different pressures.

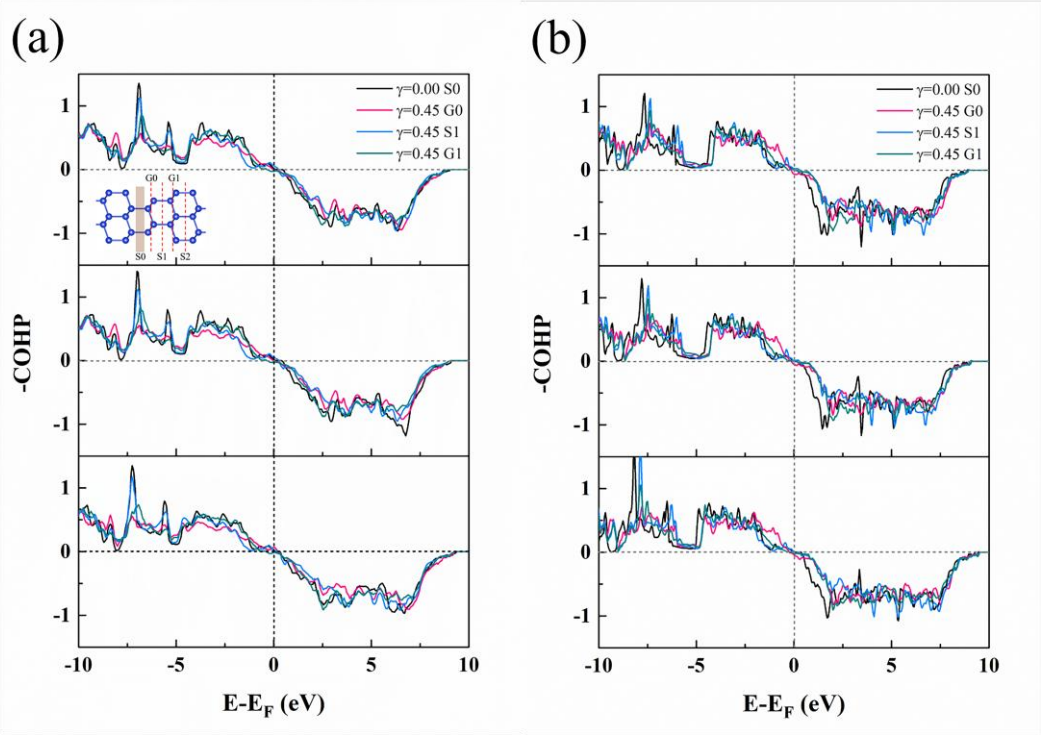

Table S1. The calculated lattice constant a (in Å), polycrystalline shear (G_{VRH}) and bulk (K_{VRH}) moduli (in GPa), Pugh ratio (G_{VRH}/B_{VRH}), ideal strength σ_{min} and τ_{min} (in GPa) under uniform strain along the weakest [111] tensile and (111)[$11\bar{2}$] shear direction, respectively, the unstable GSFEs γ_{US} (in J/m^2) and Peierls stresses τ_P (in GPa) along the [$1\bar{1}0$] and [$11\bar{2}$] directions on the shuffle-set and glide-set plane for diamond, c-BN and Si, together with the experimental data and other theoretical values for comparison.

Material	a	G_{VRH}	B_{VRH}	Pugh ratio	σ_{min}	τ_{min}	$\gamma_{US\text{-}shuffle\text{-}[1\bar{1}0]}$	$\gamma_{US\text{-}glide\text{-}[11\bar{2}]}$	$\tau_{P\text{-}shuffle\text{-}[1\bar{1}0]}$	$\tau_{P\text{-}glide\text{-}[11\bar{2}]}$
Diamond	3.535	541.34	467.85	1.157	92.33	93.2	9.97	5.77	14.3	14.8
	3.534 [1]	543.8 [1]	471.18 [1]	1.154 [1]	95 [2]	93 [2]	9.93 [1]	5.77 [3]	13.3 [3]	19.5 [3]
	3.567 [4,5]	545 [6]	468 [6]	1.165 [6]	90.7 [1]	93.9 [1,3]	9.72 [3]	5.74 [7]	13.0 [8]	
c-BN	3.583	402.73	403.22	0.999	65.4	65.3	7.47	3.76	10.83	2.83
	3.583 [1]	403.51 [1]	404.41 [1]	0.998 [1]	64.9 [3]	65.2 [3]	7.44 [3]	3.79 [3]	12.4 [3]	3.79 [3]
	3.62 [9]		400 [10]		65.9 [1]	64.9 [1]	7.20 [8]	3.43 [8]	10.3 [8]	3.73 [8]
Si	5.403	63.24	96.83	0.6531	20.97	7.25	1.45	1.82	1.3	14.28
	5.39 [11]	56.6 [12]	97.9 [13]		19.2 [8]	7.8 [8]	1.67 [14]	1.91 [14]	1.7 [8]	10-26[15]
	5.375 [12]	57.9 [11]	99.2 [8]		22 [2]	6.8 [2]		1.89 [16]	0.6-2.8 [15]	
Ge	5.65	47.12	72.35	0.6513	14.38	5.08	1.10	1.55	0.60	13.78
	5.65 [17]	46 [17]	83 [17]	0.5542 [17]	14 [2]	4.3 [2]	1.20 [18]		0.4-0.5 [18]	
	5.66 [19]	47 [19]	77 [19]	0.6104 [19]		5.4 [17]				

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