

Table S1

Due to delocalization of vibrational motion and coupling of normal modes, some cluster motions have more than one vibrational frequency associated with them. In these cases, the normal mode with the largest contribution to the relevant motion is listed in the first column

	vibration	frequency		
<i>di-</i> σ				
C ₂ H ₂ perp to dimer row		112.41		
C ₂ H ₂ parallel to dimer row		114.58	193.67	185.89
cluster breathing		149.21		
dimer seesaw		157.65		
dimer row in plane twist		193.38	257.09	
both dimers, in plane twist		531.94		
acetylene in plane torsion		341.68	359.13	
unsaturated dimer buckling		220.98		
saturated dimer buckling		243.80		
unsaturated dimer stretch		503.96		
saturated dimer stretch		508.05		
acetylene/dimer C-Si symmetric stretch		720.29		
acetylene/dimer C-Si antisymmetric stretch		753.53	760.00	
H-C-Si Bend		732.01		
acetylene C-H antisymmetric wag		1020.80		
acetylene C-H symmetric wag		766.99		
acetylene C-H symmetric wag in-plane (of acetylene)		1171.14		
acetylene C-H asymmetric wag in-plane (of acetylene)		1396.38		
C-C Stretch		1575.81		
C-H Stretch antisymmetric		3302.35		
C-H Stretch symmetric		3324.75		
<i>endbridge</i>				
C ₂ H ₂ perp to dimer row		134.62	144.98	
C ₂ H ₂ parallel to dimer row		342.72	163.86	200.6
C ₂ H ₂ parallel to dimer row + surface buckle		247.67		
symmetric buckle		261.68	458.17	
acetylene quasi in plane torsion		363.66		
acetylene quasi in plane torsion/surface buckle		435.52	475.67	
dimer stretch		479.81	499.23	
acetylene/dimer C-Si symmetric stretch		635.66		
acetylene/dimer C-Si antisymmetric stretch		739.55	760.53	
acetylene C-H antisymmetric wag		1026.27		
acetylene symmetric in-plane wag		1241.86		
acetylene antisymmetric in-plane wag		1432.08		
C-C stretch		1565.77		
C-H Stretch antisymmetric		3288.63		
C-H Stretch symmetric		3313.04		
<i>pbridge</i>				
C ₂ H ₂ parallel to dimer row		159.18	291.69	350.12
C ₂ H ₂ perp to dimer row		168.07		
surface + acetylene twist		279.70		
symmetric surface + acetylene buckle		264.38		
antisymmetric surface buckle		279.70		
acetylene/dimer C-Si symmetric stretch		581.78	535.76	602.79
acetylene/dimer C-Si antisymmetric stretch/parallel to dimer row		543.72		
acetylene/dimer C-Si antisymmetric stretch		620.83	663.43	678.58
C-C stretch		891.72		
acetylene symmetric wag		1024.11		
acetylene antisymmetric wag		1024.77	1027.36	1040.16
acetylene C-H symmetric in-plane wag		1160.83		
acetylene antisymmetric in-plane wag		1315.94		
C-H Stretch antisymmetric		3262.93		
C-H Stretch symmetric		3276.02		
<i>rbridge</i>				
asymmetric in-plane dimer slide		146.75		
symmetric in-plane dimer slide		154.13		
dimer buckle asymmetric		154.79	259.62	
dimer buckle symmetric		162.13		
C ₂ H ₂ parallel to dimer row		344.41	378.22	

C ₂ H ₂ perp to dimer row	378.80	229.04		
acetylene in plane torsion/surface buckle	528.79	584.41	433.21	462.92
acetylene/dimer C-Si symmetric stretch	450.28	460.03	641.71	
acetylene/dimer C-Si antisymmetric stretch	536.91	681.60	742.40	
acetylene/dimer C-Si antisymmetric stretch/parallel to dimer row	571.46			
C-C Stretch	946.73			
acetylene symmetric wag	991.36			
acetylene antisymmetric wag	996.92			
acetylene C-H symmetric in-plane wag	1227.64			
acetylene antisymmetric in-plane wag	1351.97			
C-H Stretch antisymmetric	3296.60			
C-H Stretch symmetric	3316.88			
cross				
acetylene moving diagonal to dimer row, perpendicular to C-C bond	146.65	203.57	206.37	
acetylene moving diagonal to dimer row, in direction of C-C bond	171.37	521.39		
dimer buckle asymmetric	269.39			
dimer buckle symmetric	294.97			
acetylene in plane torsion	395.45	443.84	364.55	398.59
Acetylene torsion, symmetric surface buckle	562.44			
acetylene out of plane wag	646.22			
C-Si antisymmetric stretch	716.63	671.02	682.88	698.51
out of plane twist	1000.94			
in plane scissoring	1242.57			
in plane rocking	1417.31			
C-C stretch	1508.84			
C-H Stretch antisymmetric	3285.83			
C-H Stretch symmetric	3308.29			