Supporting Information

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SI Methods

Fitting of Equation of States. The bulk modulus (B_0; GPa) and the equivalent volume of the carbon allotropes are obtained by least-square fitting of the equation of states (EOS). Three universal forms of EOS, namely, Vinet EOS (1), Murnaghan EOS (2), and the third-order Birch-Murnaghan EOS (3), are independently used for the fitting, all of which yield consistent results.

Vinet EOS.

$$P(V)=3B_0x^{-2}(1-x)\exp[\eta(1-x)],\quad x=\left(\frac{V}{V_0}\right)^{1/3},\quad \eta=\frac{3}{2}(B_0'-1).$$

Murnaghan EOS.

$$P(V)=\frac{B_0}{B_0'}\left[\left(\frac{V_0}{V}\right)^{B_0'}-1\right].$$

Third-Order Birch-Murnaghan EOS.

$$P(V)=\frac{3B_0}{2}\left(x^{-7}-x^{-5}\right)\left[1+\frac{3}{4}(B_0'-4)\left(x^{-2}-1\right)\right],\quad x=\left(\frac{V}{V_0}\right)^{1/3}.$$

Here P and V denote the pressure and the corresponding volume of cell, respectively. V_0 is the equivalent volume at zero pressure and B_0' is the bulk modulus pressure derivative. Integrating these two EOSs with V yields the corresponding E-V relationship, where E is the total energy at volume V.

Vinet EOS.

$$E(V)=E_0+\frac{9B_0V_0}{\eta^2}\left\{1-[1-\eta(1-x)]\exp[\eta(1-x)]\right\}.$$

Murnaghan EOS.

$$E(V)=E(0)+\frac{B_0V}{B_0'}\left[\frac{(V_0/V)^{B_0'}}{B_0'-1}+1\right]-\frac{B_0V_0}{B_0'-1}.$$

Third-Order Birch-Murnaghan EOS.

$$E(V)=E_0+\frac{9B_0V_0}{16}\left[\left(x^{-2}-1\right)^3B_0'+\left(x^{-2}-1\right)^2\left(6-4x^{-2}\right)\right],$$
$$x=\left(\frac{V}{V_0}\right)^{1/3}.$$

1. Vinet P, Rose JH, Ferrante J, Smith JR (1989) Universal features of the equation of state of solids. *J Phys Condens Matter* 1(11):1941.
2. Murnaghan FD (1944) The compressibility of media under extreme pressures. *Proc Natl Acad Sci USA* 30(9):244–247.
3. Birch F (1947) Finite elastic strain of cubic crystals. *Phys Rev* 71(11):809–824.

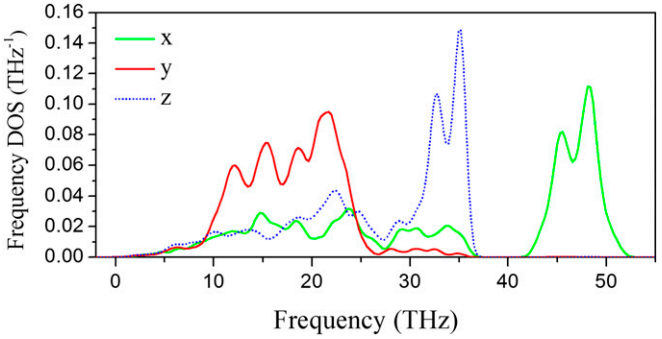

Fig. S1. Direction-resolved phonon frequency density of states for C2 atoms in T6-carbon. The *x* direction here is parallel to the C=C double bond in the structure.

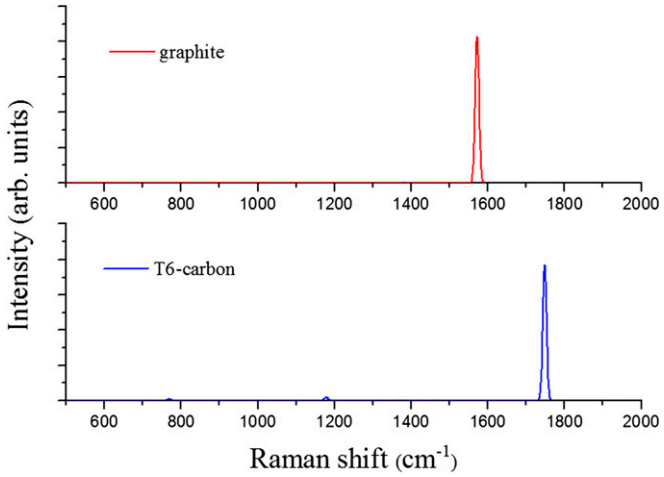

Fig. S2. Calculated first-order Raman spectra of graphite and T6-carbon. Our result in graphite is in good agreement with previous work (1).

1. Tuinstra F, Koenig JL (1970) Raman spectrum of graphite. *J Chem Phys* 53(3):1126–1130.

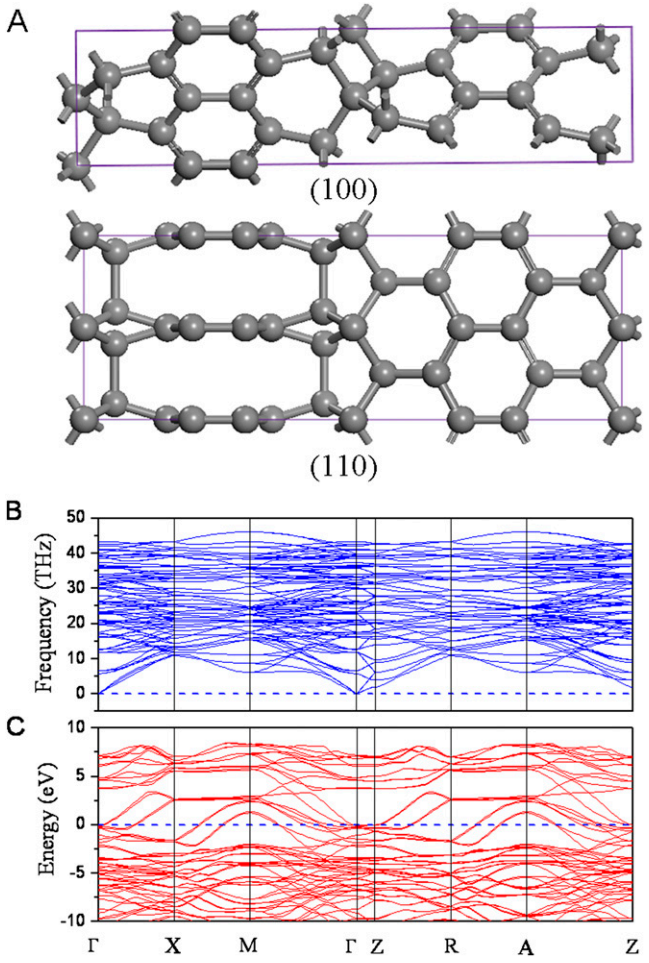

Fig. S3. (*A*) Optimized geometry of T28-carbon viewed along the [100] and [110] directions, (*B*) phonon dispersions, and (*C*) electronic band structure of T28-carbon.

Table S1. Optimized structural data of the carbon allotropes investigated in this work

Structure	Lattice parameters (Å) a	b	c	Atomic Wyckoff positions
T6	2.60		6.00	C(2*f*): (1/2, 1/2, 1/4)
$P4_2/mmc$ (131)				C(4*i*): (0, 1/2, 0.1118)
T14	2.56		14.62	C(4*h*): (1/2, 1/2, 0.9517)
$P4_2/mmc$ (131)				C(4*i*): (1/2, 0, 0.9027)
				C(4*i*): (1/2, 0, 0.8084)
				C(2*f*): (1/2, 1/2, 1/4)
T12 (1)	3.43		6.09	C(8*i*): (0.3319, 0.8319, 0.3597)
$P4_2/ncm$ (138)				C(4*b*): (0, 0, 0)
T28	3.55		14.66	C(8*i*): (0.2478, 0.2522, 0.6539)
$P4_2/ncm$ (138)				C(8*i*): (0.8385, 0.6615, 0.5559)
				C(8*f*): (0, 0, 0.2991)
				C(4*b*): (0, 0, 0.5)
Cubic-C_{16} (2)	4.88			C(16*f*): (0.3374, 0.3374, 0.3374)
$Im\bar{3}m$ (229)				
T-carbon (3)	7.52			C(32*e*): (0.0706, 0.0706, 0.0706)
$Fd\bar{3}m$ (227)				
L-carbon (4)	5.32		8.68	C(12*k*): (0.4274, 0.8548, 0.5267)
$P6_3/mmc$ (194)				C(4*f*): (1/3, 2/3, 0.6681)
Y-carbon (5)	9.64			C(8*a*): (0, 0, 0)
$Fd\bar{3}m$ (227)				C(32*e*): (0.0888, 0.0888, 0.0888)
TY-carbon (5)	13.46			C(32*e*): (0.0397, 0.0397, 0.0397)
$Fd\bar{3}m$ (227)				C(32*e*): (0.0987, 0.0987, 0.0987)

1. Zhao Z, et al. (2012) Tetragonal allotrope of group 14 elements. *J Am Chem Soc* 134(30):12362–12365.
2. Srinivasu K, Ghosh SK (2012) Electronic structure, optical properties, and hydrogen adsorption characteristics of supercubane-based three-dimensional porous carbon. *J Phys Chem C* 116(47):25015–25021.
3. Sheng X-L, Yan Q-B, Ye F, Zheng Q-R, Su G (2011) T-carbon: A novel carbon allotrope. *Phys Rev Lett* 106(15):155703.
4. Yang L, He HY, Pan BC (2013) Theoretical prediction of new carbon allotropes. *J Chem Phys* 138(2):024502–024506.
5. Jo JY, Kim BG (2012) Carbon allotropes with triple bond predicted by first-principle calculation: Triple bond modified diamond and T-carbon. *Phys Rev B* 86(7):075151.

Table S2. Calculated equilibrium volume (V_0), bulk modulus (B_0), and corresponding pressure derivatives (B_0')

Structure	ρ (g/cm^3)	B_0 (GPa)	B_0'	V_0 (Å^3/atom)
Diamond	3.550	433.1 (1) (454.6)	3.71	5.621
Lonsdaleite	3.487	433.1 (1) (454.5)	3.63	5.719
graphite	2.017	245.0	3.51	9.890
T6	2.952	337.4	3.79	6.766
T12	3.342	403.2 (1) (424.8)	3.75	5.968
T14	2.923	349.6	3.69	6.800
T28	3.019	356.2	3.46	6.606
Cubic-C_{16}	2.750	309.1	3.74	7.253
T-carbon	1.503	159.8 (2) (169)	3.61	13.274
L-carbon	1.500	158.1 (3) (159)	3.80	13.293
Y-carbon	0.891	83.5 (4) (82.9)	3.74	22.373
TY-carbon	0.523	54.3 (4) (54.2)	3.71	38.103

The numbers in parentheses are also listed from the given references for comparison.

1. Zhao Z, et al. (2012) Tetragonal allotrope of group 14 elements. *J Am Chem Soc* 134(30):12362–12365.
2. Sheng X-L, Yan Q-B, Ye F, Zheng Q-R, Su G (2011) T-carbon: A novel carbon allotrope. *Phys Rev Lett* 106(15):155703.
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