Supporting Information

This article mainly supplements some details and contents of the main text. Including the structure optimization of RDX; the variation of partial chemical bonds of RDX deviating from the equilibrium bond length at different temperatures; the mathematical details of calculating the maximum gravitational position of chemical bonds; the maximum gravitational position of each chemical bond of RDX; the structure optimization of nitromethane; nitromethane under different pressures the maximum gravitational position of each chemical bond; the vibration of hydrogen atoms in all directions; the variation of each chemical bond of nitromethane deviating from the equilibrium bond length under different pressures and temperatures.

Structural optimization of RDX. Figure S1(a) shows the optimized crystal structure of α-RDX at 0GPa, with 8 RDX molecules in each unit cell. The structure of a single RDX molecule is shown in Figure S1(b), and the molecular configuration of RDX is the AAE configuration, which is consistent with the experiment. As shown in Table S1, the optimized lattice parameters are also in good agreement with the experimental lattice parameters.

The variation of the chemical bonds of the RDX part deviating from the equilibrium bond length at different temperatures. As shown in Figure S2, with the increase of temperature, the variation of each chemical bond of RDX deviating from the equilibrium bond length will increase.

Mathematical details for calculating the position of maximum gravitational force in chemical bonds. This work uses the energy curve to derive the displacement, and

the point where the first derivative is equal to zero is the equilibrium position of the chemical bond. Let the point where the second derivative of the energy curve with respect to the displacement equals zero be the position of maximum gravitational force of the chemical bond. The detailed formula for the derivation is as follows:

$$f^{'}(x)=\lim_{h\to 0}\frac{f(x+h)-f(x)}{h}$$

When h is small enough, we make the following approximation:

$$f^{'}(x)\approx\frac{f(x+h)-f(x-h)}{2h}$$

The maximum gravitational position of each chemical bond in RDX. As shown in Table S2, the maximum gravitational position R_m of each chemical bond of α-RDX under 0GPa, the equilibrium position R_0 and the tensile variable Δ of the theoretical bond breaking of the chemical bond are. Where N-NO_2 is the maximal gravitational position of the N-N bond when one of the oxygen atoms on the nitro group is about to fall off.

Structural optimization of nitromethane. Figure S3(a) below shows the optimized crystal structure of nitromethane. Each protocell consists of four nitromethane molecules with space group $P2_12_12_1$. Figure S3(b) shows the optimized single nitromethane molecule at 12GPa. It can be seen from the figure that the two hydrogens on the methyl group and the two oxygens on the nitro group are in the same plane, which is consistent with other studies at this pressure. The conclusion that the lower nitromethane molecule is in an overlapping conformation is consistent. At the same time, we also compare the calculated lattice parameters with other studies [40, 41] as shown in Table S3. From the table, we can see that the lattice parameters calculated by

us are basically consistent with the lattice parameters obtained by the experiment, which indicates the rationality of the calculation results of this work.

The maximum gravitational position of each chemical bond in nitromethane at different pressures. As shown in Table S4, the maximum gravitational positions of the chemical bonds are basically unchanged over the small pressure ranges studied. Where C-NO_2 is the maximal gravitational position of the C-N bond when one of the oxygen atoms on the nitro group is about to fall off.

Vibration of hydrogen atoms in all directions. As shown in Figure S4 below, the vibrations of hydrogen atoms in all directions are shaded. It can be seen from the figure that the vibration of hydrogen atoms in all directions is displayed in the form of an ellipsoid. Obviously, we cannot regard the vibrations of the hydrogen atoms in nitromethane as anisotropic.

The variation of each chemical bond of nitromethane deviating from the equilibrium bond length at different pressures and temperatures. As shown in Figure S5, the stretching amount of chemical bonds increases with the increase of temperature under the same pressure. At the same temperature, the amount of stretching of chemical bonds decreases with increasing pressure.

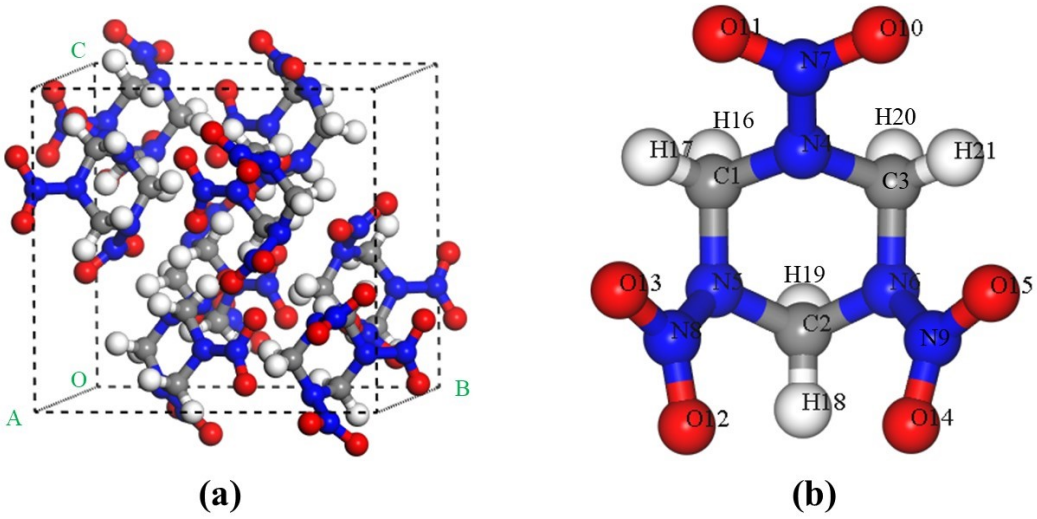

Fig. S1. Crystal Structure and Molecular Structure of α-RDX at 0GPa

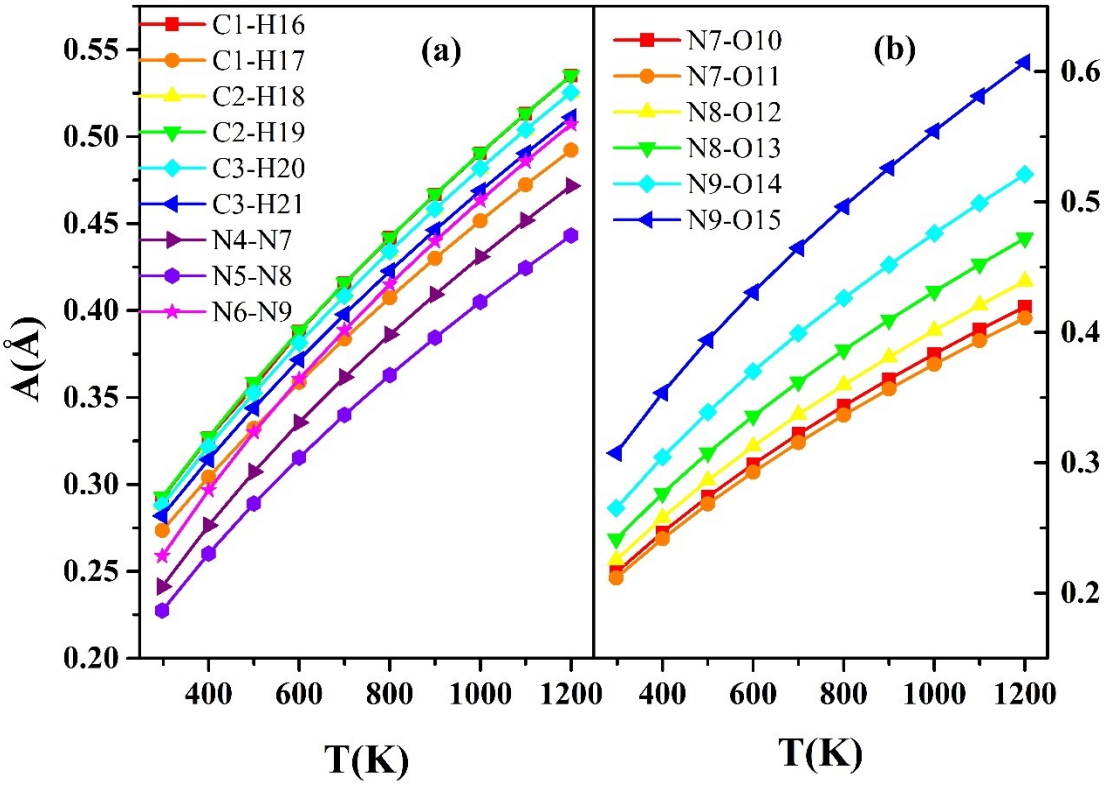

Fig. S2. The stretching amount of each chemical bond in RDX deviating from the equilibrium bond length as a function of temperature at 0GPa

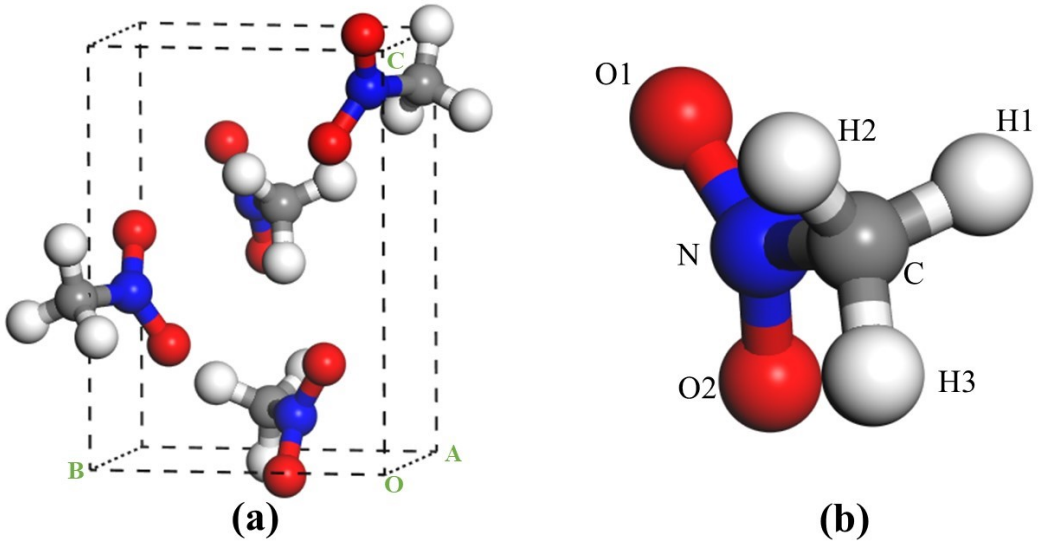

Fig. S3. Crystal structure and molecular structure of nitromethane

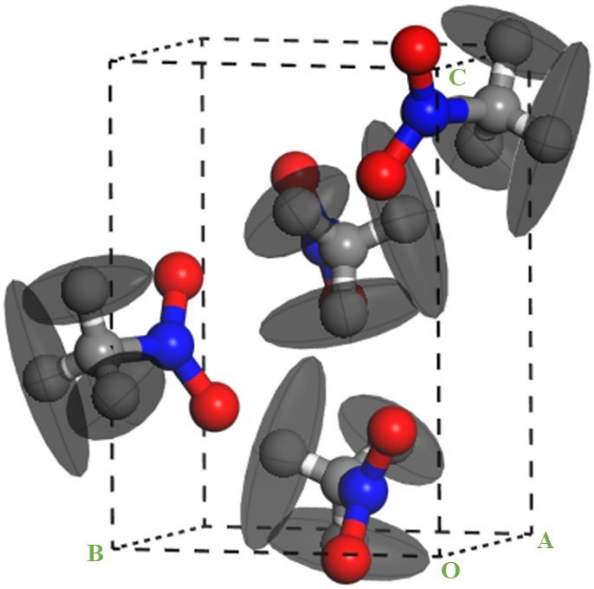

Fig. S4. Mean square displacement of hydrogen atomic anisotropy in nitromethane

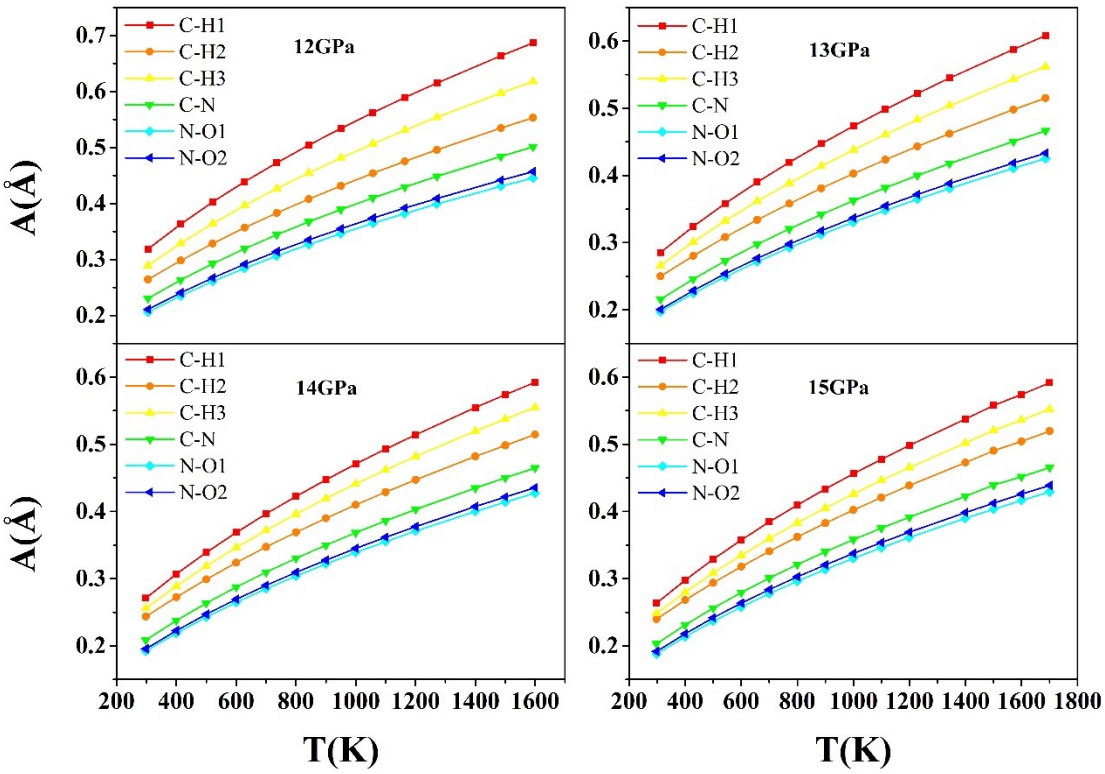

Fig. S5. Offset of each bond from equilibrium bond length in solid nitromethane at different pressures and temperatures

Table S1 Experimental and calculated lattice parameters of α-RDX at 0GPa.

a/Å	b/Å	c/Å	V/Å^3	Reference
13.3472	11.5061	10.7575	1652.07	This work
13.182	11.574	10.709	1633.86	Expt. [32]

Table S2 The maximum gravitational position Rm of each chemical bond of α-RDX under 0GPa, the equilibrium position R0 and the stretching variable of the theoretical bond breaking.

Bond	R_m/Å	R_0/Å	Δ/Å
C1-H16	1.504495	1.1016	0.402895
C1-H17	1.53471	1.09132	0.44339
C2-H18	1.544985	1.09064	0.454345
C2-H19	1.514955	1.09512	0.419835
C3-H20	1.504485	1.10073	0.403755
C3-H21	1.53499	1.09072	0.44427
N4-N7	1.76485	1.39502	0.36983
N5-N8	1.81506	1.44446	0.3706
N6-N9	1.845095	1.45045	0.394645
N7-O10	1.55491	1.24321	0.3117
N7-O11	1.554765	1.23819	0.316575
N8-O12	1.544755	1.22953	0.315225
N8-O13	1.545125	1.23392	0.311205
N9-O14	1.544945	1.23117	0.313775
N9-O15	1.545	1.22911	0.31589
$N4\text{-}N7O_2$	1.664875	1.39502	0.269855
$N5\text{-}N8O_2$	1.714555	1.44446	0.270095
$N6\text{-}N9O_2$	1.715	1.45045	0.26455

Table S3 Experimental and calculated lattice parameters for nitromethane.

Condition	a/Å	b/Å	c/Å	V/Å^3	Reference
0K 0GPa	5.20027	6.36860	8.48329	280.954	This Work
4.8k 0.00001GPa	5.1832	6.2357	8.5181	275.307	Expt. [40]
78K 0.00001GPa	5.1983	6.2457	8.5640	278.048	Expt. [40]
0K 15GPa	4.39958	5.43828	7.69491	184.110	This Work
298K 15GPa	4.540	5.534	7.687	193.131	Expt. [41]

Table S4 The equilibrium position R_0 of each chemical bond of nitromethane under different pressures, the maximum gravitational position R_m and the tensile change Δ of the theoretical bond breaking.

Pressure	Bond	R_m/Å	R_0/Å	Δ/Å
12GPa NM	C-H1	1.53539	1.08513	0.45026
	C-H2	1.524115	1.08166	0.442455
	C-H3	1.52478	1.08337	0.44141
	C-N	1.91542	1.46818	0.44724
	N-O1	1.57516	1.2347	0.34046
	N-O2	1.585635	1.23083	0.354805
	C-NO_2	1.865225	1.46818	0.397045
13GPa NM	C-H1	1.535625	1.08455	0.451075
	C-H2	1.524955	1.08118	0.443775
	C-H3	1.52524	1.08279	0.44245
	C-N	1.91583	1.4663	0.44953
	N-O1	1.575175	1.23427	0.340905
	N-O2	1.584695	1.23047	0.354225
	C-NO_2	1.87014	1.4663	0.40384
14GPa NM	C-H1	1.53561	1.08397	0.45164
	C-H2	1.535225	1.0806	0.454625
	C-H3	1.52552	1.08231	0.44321
	C-N	1.915045	1.4644	0.450645
	N-O1	1.574415	1.23396	0.340455
	N-O2	1.58476	1.22987	0.35489
	C-NO_2	1.844905	1.4644	0.380505
15GPa NM	C-H1	1.53551	1.08351	0.452
	C-H2	1.52591	1.07993	0.44598
	C-H3	1.52369	1.08171	0.44198
	C-N	1.91507	1.46255	0.45252
	N-O1	1.57502	1.23343	0.34159
	N-O2	1.58397	1.22948	0.35449
	C-NO_2	1.854875	1.46255	0.392325