Supplementary Information (SI) for Journal of Materials Chemistry C.

Structural phase transition, mechanics, and thermodynamics of heavy fermion metal UPt_3 under pressure

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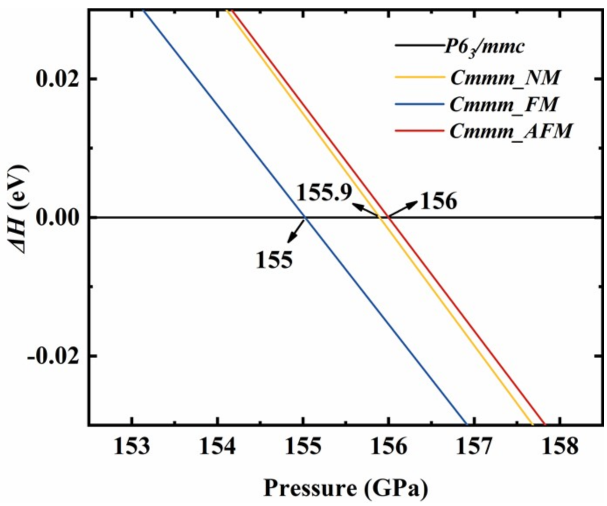

Fig. S1 Effect of different magnetic states on the P_T of *$P6_3/mmc$* → *Cmmm*. The enthalpy of the *$P6_3/mmc$* phase is set as zero for reference.

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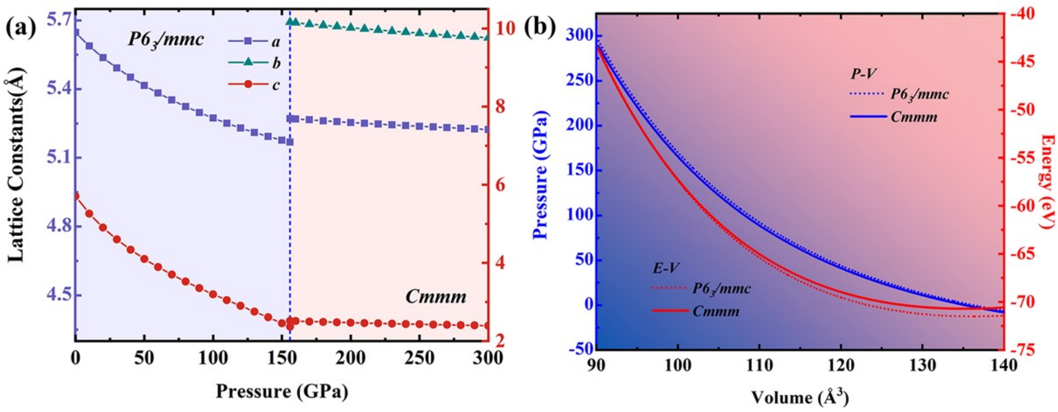

Fig. S2 (a) Dependences of lattice constants on pressure. (b)Total energy (*E*)–volume (*V*) and the pressure (*P*)–*V* relations of UPt_3 by EOS fitting at 0 GPa.

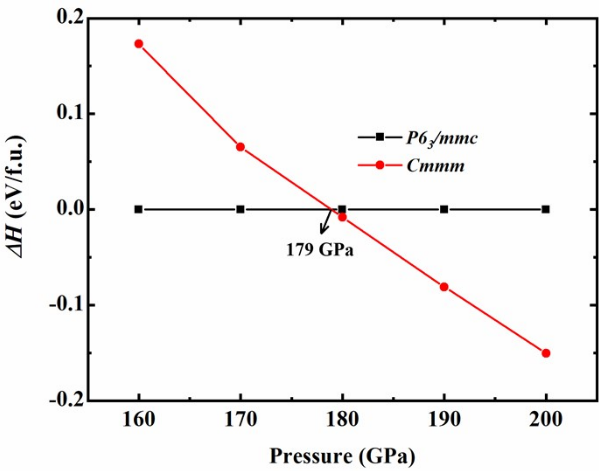

Fig. S3 Enthalpy difference diagram included the SOC effect.

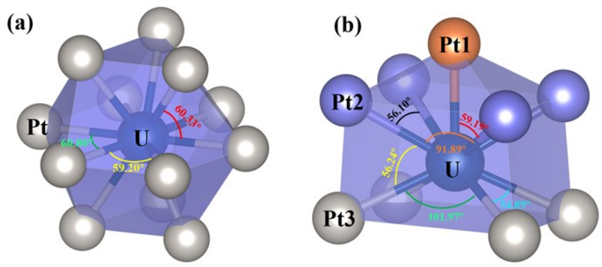

Fig. S4 Bond angles of (a) *$P6_3/mmc$* and (b) *Cmmm* UPt_3 at 155.9 GPa.

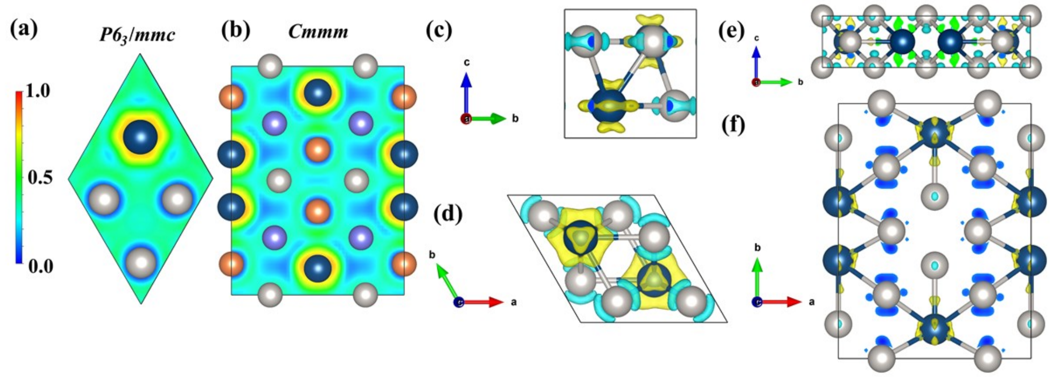

Fig. S5 (a)-(b) ELFs and (c)-(f) differential charge density of *$P6_3/mmc$*-UPt_3 and *Cmmm*-UPt_3 at 155.9 GPa. The yellow color (isosurface = +0.001 e/bohr^3) indicates the loss of electrons from U atoms and the blue color (isosurface = -0.09 e/bohr^3) represents the grain of electrons between Pt atoms.

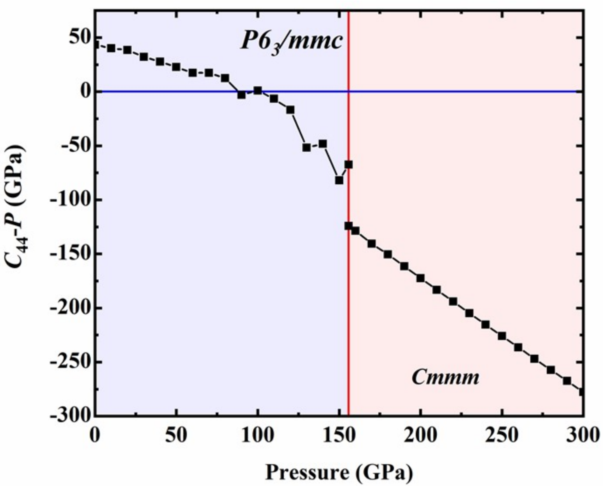

Fig. S6 Modified Born stability conditions under pressure for UPt_3

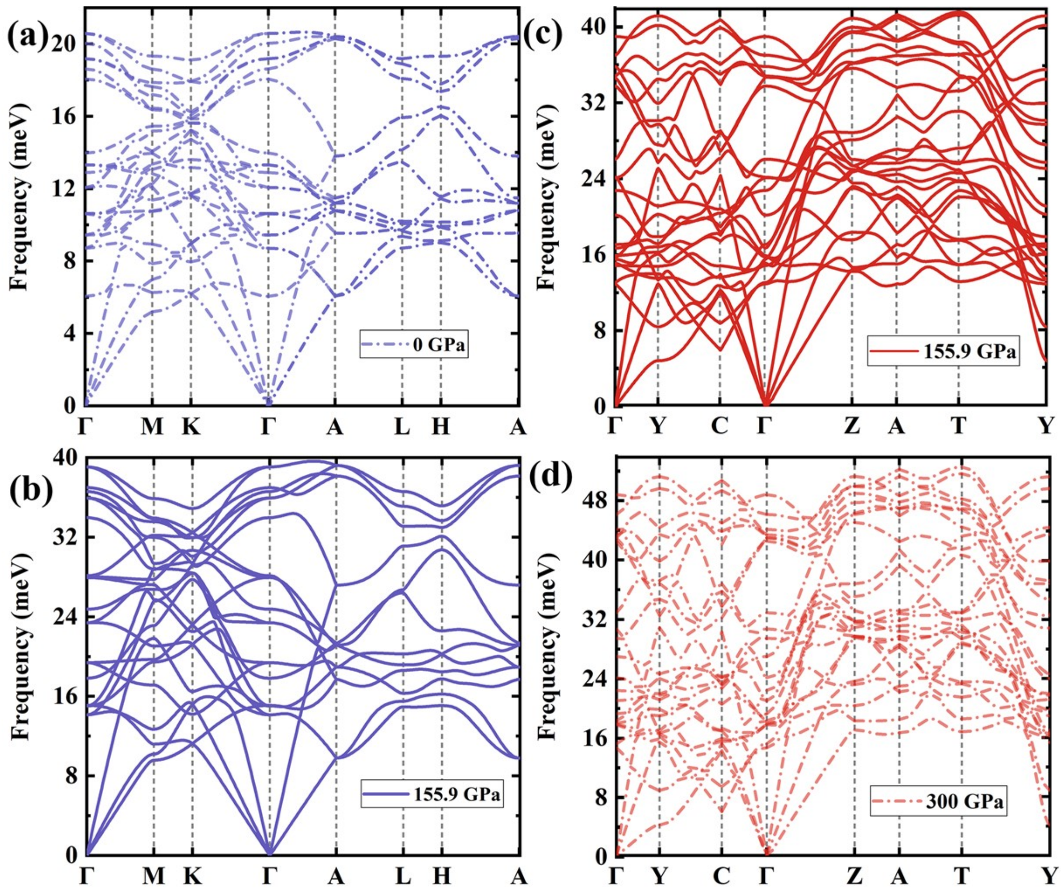

Fig. S7 Calculated phonon dispersions for (a)-(b) $P6_3/mmc$ and (c)-(d) $Cmmm$ phases at different pressures.

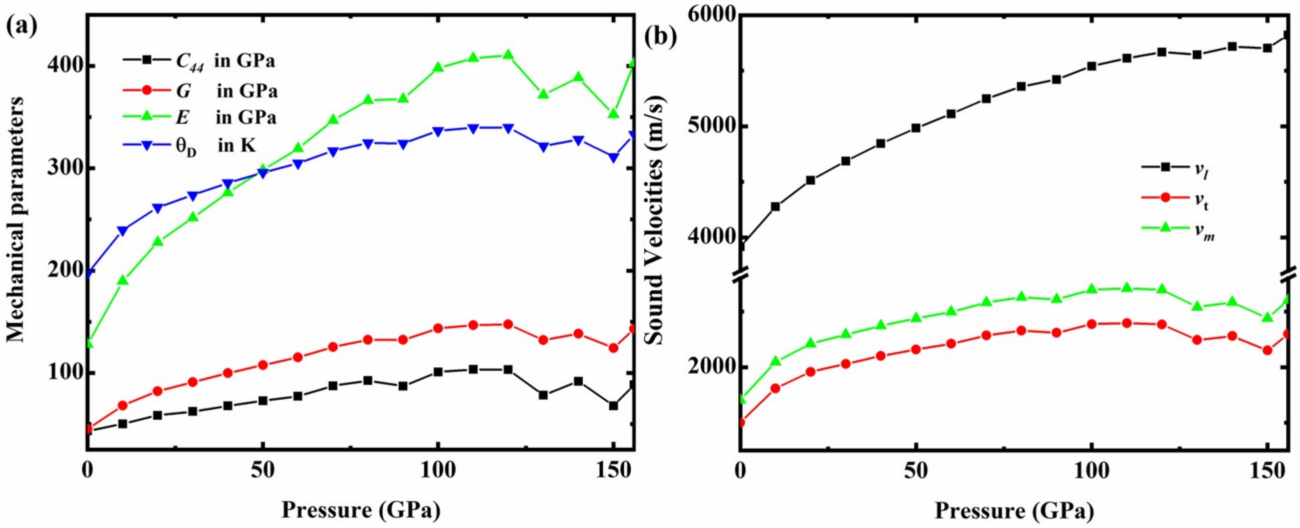

Fig. S8 Calculated (a) mechanical parameters and (b) sound velocities as a function of pressure for *P6₃/mmc*-UPt₃.

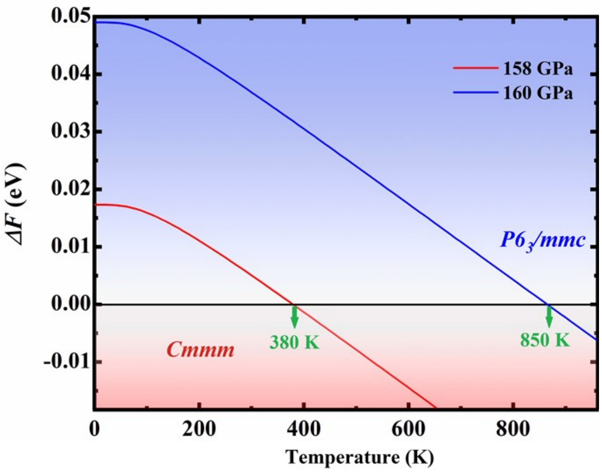

Fig. S9 Relative free energy(ΔF) as a function of temperature.

Table. S1 Volume V_0 (Å^3), energy (eV), bulk modulus B_0 (GPa), and pressure derivative of the bulk modulus B' at 0 GPa for EOS fitting.

Space group	V_0	E_0	B_0	B'
$P6_3/mmc$	136.25	-71.50	250.79	5.04
$Cmmm$	135.23	-70.70	254.53	5.06

Table. S2 At 155.9 GPa, Infrared (IR) and Raman (R) active optical phonon modes and their irreducible representations (IRREP) at Γ-point for *P6₃/mmc* and *Cmmm* phases. The unit of phonon frequency is in meV.

	$P6_3/mmc$			$Cmmm$	
IRREP	Frequency	Activity	IRREP	Frequency	Activity
E_{2g}	14.14	R	B_{3g}	12.93	R
			B_{1u}	13.07	IR
E_{2g}	15.03	R	B_{2g}	14.86	R
			B_{3g}	15.13	R
B_{2g}	15.09		B_{2u}	15.51	IR
A_{2g}	17.83		B_{1u}	15.75	IR
E_{1g}	19.39	R	A_u	15.92	
			B_{1g}	16.70	R
E_{1u}	23.40	IR	A_g	17.08	R
			B_{3u}	20.16	IR
B_{2u}	24.76		B_{1u}	22.76	IR
E_{2u}	27.93		B_{2u}	24.10	IR
			B_{1g}	24.15	R
A_{2u}	28.13	IR	B_{3u}	26.06	IR
B_{1u}	33.96		B_{3u}	33.75	IR
E_{1u}	35.92	IR	B_{2u}	34.67	IR
			A_g	34.79	R
A_{1g}	36.58	R	B_{1g}	34.82	R
B_{2g}	36.97		B_{2u}	35.81	IR
E_{2g}	39.05	R	A_g	37.00	R
			B_{3u}	39.7	IR

The polycrystalline bulk modulus B, shear modulus G, Young's modulus E, and Poisson's ratio (σ) of UPt_3 are estimated using the Voigt-Reuss-Hill (VRH) approximations. They are defined as following: $B = \frac{B_V + B_R}{2}$, $G = \frac{G_V + G_R}{2}$, $E = \frac{9BG}{3B + G}$ and $\sigma = \frac{3B - 2G}{2(3B + G)}$. For a hexagonal system, detailed representations of B and G are:

$$B_V = \frac{2(C_{11} + C_{12}) + C_{33} + 4C_{13}}{9}, \tag{1}$$

$$B_R = \frac{(C_{11} + C_{12})C_{33} + 2C_{13}^2}{C_{11} + C_{12} + 2C_{33} - 4C_{13}}, \tag{2}$$

$$G_V = \frac{7C_{11} + 5C_{12} + 12C_{44} + 2C_{33} - 4C_{13}}{30}, \tag{3}$$

$$G_R = \frac{5[(C_{11} + C_{12})C_{33} - 2C_{13}^2]^2 C_{44} C_{66}}{6B_V C_{44} C_{66} + 2[(C_{11} + C_{12})C_{33} - 2C_{13}^2]^2 (C_{44} + C_{66})}. \tag{4}$$

The θ_D and elastic wave velocities are related to the elastic moduli in relations to:

$$\theta_D = \frac{h}{k_B}[\frac{3nN_A\rho}{4\pi M}]^{1/3} v_m, \quad v_m = [\frac{1}{3}(\frac{2}{v_t^3} + \frac{1}{v_l^3})]^{-1/3}, \quad v_t = \sqrt{G/\rho}, \quad v_l = \sqrt{(3B + 4G)/3\rho}, \tag{5}$$