Supplemental Material for "Superconducting and structural properties of the type-I superconductor $PdTe_2$ under high pressure"

Yusaku Furue,[1] Tadachika Fujino,[2] Marc V. Salis,[3] Huaqian Leng,[3,4] Fumihiro Ishikawa,[2] Takashi Naka,[5] Satoshi Nakano,[5] Yingkai Huang,[3] Anne de Visser,[3] and Ayako Ohmura[2,*]

[1] *Graduate School of Science and Technology, Niigata University, 8050, Ikarashi 2-no-cho, Nishi-ku, Niigata, 950-2181, Japan*

[2] *Faculty of Science, Niigata University, 8050, Ikarashi 2-no-cho, Nishi-ku, Niigata, 950-2181, Japan*

[3] *Van der Waals-Zeeman Institute, University of Amsterdam, Science Park 904, 1098 XH Amsterdam, The Netherlands*

[4] *School of Physics, University of Electronic Science and Technology of China, Chengdu, China, 610054*

[5] *National Institute for Materials Science (NIMS), Sengen 1-2-1, Tsukuba, Ibaraki 305-0047, Japan*

1. Rietveld analysis of x-ray diffraction patterns and the list of structural parameters of $PdTe_2$

The Rietveld analysis was performed using the crystallographic program JANA2006 in order to obtain the structural parameter [1, 2]. Figure S1 shows a typical result for one of the diffraction patterns measured at $P = 0$ GPa. It demonstrates the quality of the fit. All diffraction patterns obtained in this study were analyzed with a similar accuracy. Table S1 shows the list of structural parameters obtained at each pressure. Values in this table were obtained in the first run with a 4:1 mixture of Methanol (MeOH)-Ethanol (EtOH) used as pressure transmitting medium. Since the MeOH-EtOH medium maintains a liquid condition up to 10 GPa, the hydrostaticity in the sample space can be kept at the maximum pressure of 8 GPa in this study. The lattice parameters a(Å), c(Å), and the unit cell volume V(Å^3), monotonically decrease with pressure. The z-position of Te (z_{Te}) remains constant within the error up to ~ 1 GPa and then starts to increase as a function of pressure.

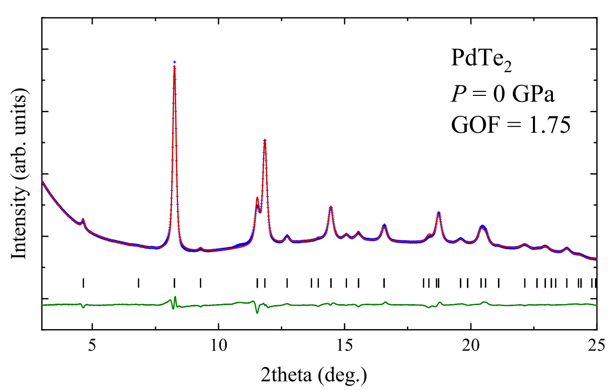

FIG. S1. X-ray diffraction pattern of $PdTe_2$ measured at $P = 0$ GPa and the Rietveld refinement fitting results. Blue, red, and green lines indicate the experimental and calculated diffraction intensities, as well as the residual error. Small bars below the diffraction pattern indicate the diffraction angles in the $P\overline{3}m1$ structure. GOF means goodness of fit.

* E-mail: ohmura@phys.sc.niigata-u.ac.jp

TABLE S1. Structural parameters obtained at each pressure: lattice constants a(Å) and c(Å), volume V(Å^3) of the unit cell, and the atomic z-position of Te (z_{Te}). In the $P\overline{3}m1$ structure for $PdTe_2$ (Z=2), the atomic positions are (0, 0, 0) for Pd and (1/3, 2/3, z_{Te}) for Te. As regards the atomic position, only the value of z_{Te} can be optimized by the Rietveld analysis.

Pressure (GPa)	a (Å)	c (Å)	V (Å^3)	z_{Te}
0	4.0441(1)	5.1511(4)	72.957(5)	0.2747(5)
0.29	4.0396(1)	5.1395(4)	72.634(5)	0.2740(5)
0.62	4.0346(1)	5.1254(4)	72.255(5)	0.2741(5)
0.91	4.0305(1)	5.1139(4)	71.947(5)	0.2745(5)
1.20	4.0260(1)	5.1012(4)	71.607(5)	0.2746(5)
1.54	4.0219(1)	5.0899(4)	71.304(5)	0.2752(5)
1.93	4.0169(1)	5.0768(4)	70.942(5)	0.2756(5)
2.26	4.0123(1)	5.0652(4)	70.617(5)	0.2762(5)
2.63	4.0079(1)	5.0542(4)	70.311(5)	0.2767(5)
2.87	4.0048(1)	5.0463(4)	70.091(5)	0.2771(5)
3.21	4.0004(1)	5.0361(4)	69.796(5)	0.2777(5)
3.39	3.9980(1)	5.0301(4)	69.628(5)	0.2780(5)
3.89	3.9927(1)	5.0184(4)	69.283(4)	0.2787(5)
4.34	3.9870(1)	5.0062(4)	68.917(4)	0.2795(5)
4.90	3.9806(1)	4.9922(4)	68.506(4)	0.2799(5)
5.40	3.9752(1)	4.9823(4)	68.185(4)	0.2803(5)
5.99	3.9680(1)	4.9672(4)	67.731(4)	0.2813(5)
6.46	3.9625(1)	4.9566(4)	67.398(4)	0.2820(5)
6.91	3.9580(1)	4.9484(3)	67.137(4)	0.2826(5)
7.47	3.9512(1)	4.9362(3)	66.738(4)	0.2835(5)
7.95	3.9468(1)	4.9281(3)	66.480(4)	0.2845(5)

2. Estimation of the Debye temperature Θ_D utilizing the Bloch-Grüneisen formula

We estimated the pressure variation of the Debye temperature Θ_D from the Bloch-Grüneisen (BG) formula. The $\rho(T)$ data can be fitted to the BG formula based on an electron-phonon scattering with temperature exponent n=5:

$$\rho(T) = \rho_0 + 4.225\rho_\Theta \left(\frac{T}{\Theta_D}\right)^5 \int_0^{\Theta_D/T} \frac{x^5}{(e^x-1)(1-e^{-1})} dx,$$

where ρ_0 is the residual resistivity, Θ_D is the Debye temperature, and ρ_Θ is the resistivity at Θ_D. Besides for sample 2, which is introduced in the main manuscript, we fitted $\rho(T)$ of another sample (sample 4), which was measured using a piston-cylinder cell in the PPMS, to obtain values of Θ_D below 2.5 GPa in detail. Figure S2 shows the fitting result at 0.4 GPa for sample 4. It results in Θ_D = 184.8(4) K. Other data of samples 2 and 4 were also fitted to the BG formula with the same fitting quality. Figure S3 shows the pressure variation of Θ_D obtained for samples 2 (open symbols) and 4 (closed symbols). From the results of sample 4, the value of Θ_D seems to remain almost constant at first and then rapidly increases beyond 1 GPa. The rapid increase of Θ_D between 1 and 3 GPa is also obtained for sample 2.

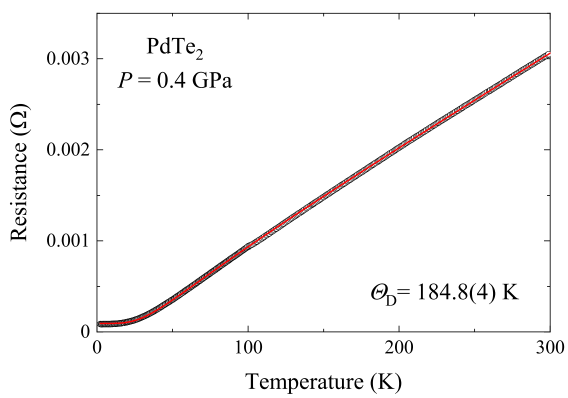

FIG. S2. The result of Bloch-Grüneisen fitting on the resistance of sample 4 measured at 0.4 GPa. The dimension of sample 4 is $\sim 1.4(W)\times2.7(L)\times0.05(H)$ mm^3 used in the piston-cylinder cell. The black symbols and red line indicate the experimental data and fitting result, respectively.

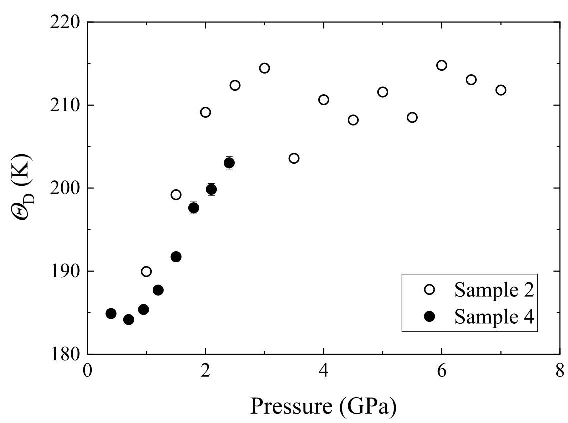

FIG. S3. Pressure variations of the Debye temperature Θ_D estimated from the experimental resistivity data. Open and closed circles indicate values of Θ_D obtained on samples 2 and 4, respectively.

3. Pressure variation of the electrical resistivity of $PdTe_2$ at room temperature

Figure S4 shows the value of the electrical resistivity at room temperature obtained for samples 1 and 2 as a function of pressure. The observed pressure variations disagree with each other, that is to say, the resistivity of sample 1 monotonically decreases with pressure, while that of sample 2 changes the tendency from downward to upward between 1 and 2 GPa. As presented in the main manuscript, the appearance of superconductivity in samples 1 and 2 is also different as shown in Fig. 5 (a) and (b). We infer that sample 1 contains a small amount of elemental Te. This possibly also affects the room temperature resistance values.

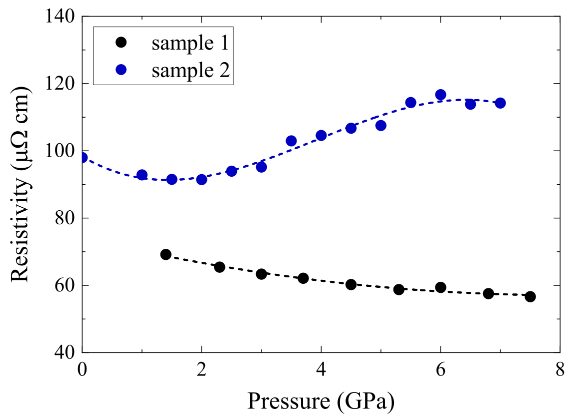

FIG. S4. Pressure variation of the electrical resistivities of samples 1 and 2 measured at room temperature. Black and blue circles indicate values obtained for samples 1 and 2, respectively. Broken lines are guides for the eye.

4. Estimation of the logarithmic volume derivative of the Hopfield parameter η

Referring to Ref. [3], the logarithmic volume derivative of T_c derived from the Bardeen-Cooper-Schrieffer (BCS) expression is shown as follows:

$$\frac{d\ln T_c}{d\ln V} = -B_0\frac{d\ln T_c}{dP} = -\frac{B_0}{T_c}\frac{dT_c}{dP} = -\gamma + \left[\ln\left(\frac{\Theta_D}{T_c}\right)\right]\left[\frac{d\ln\eta}{d\ln V} + 2\gamma\right],$$

where T_c is the superconducting transition temperature, V is the volume, B_0 is the bulk modulus, P is in units of GPa, η $(= N(E_f)\langle I^2\rangle)$ is the Hopfield parameter, γ $(= -d\ln\langle\omega\rangle/d\ln V)$ is the Grüneisen parameter, and Θ_D $(= \langle\omega\rangle/0.83)$ is the Debye temperature. The sign of dT_c/dP is expected to become positive when the logarithmic volume derivative of η, $d\ln\eta/d\ln V$, is larger in magnitude than 2γ.

In the pressure range up to 2.5 GPa, we estimated the value of $d\ln\eta/d\ln V$ below and above 1 GPa utilizing the experimental parameters, T_c, B_0, and Θ_D, obtained in this study and Ref. [4]. γ was estimated from the pressure variation of Θ_D obtained on sample 4 (see Fig. S3): $\gamma \sim 0$ at $P < 1$ GPa and ~ 3.3 at $P > 1$ GPa. The calculated value for $\ln(\Theta_D/T_c)$ was ~ 4.6 at 0.4 GPa and ~ 5.0 at 2.4 GPa. Combining these values with $B_0 = 62.9$ GPa and $d\ln T_c/dP$, the value of $d\ln\eta/d\ln V$ below 2.5 GPa can be estimated to be ~ -2.9 at $P < 1$ GPa and ~ -2.8 at $P > 1$ GPa, respectively.

[1] H. M. Rietveld, Acta Cryst., **22**, 151 (1967).

[2] JANA2006, Crystallographic Computing System, http://jana.fzu.cz/

[3] J. S. Schilling, in *Handbook of High-Temperature Superconductivity: Theory and Experiment*, edited by J. R. Schrieffer and J. S. Brooks (Springer, New York, 2007).

[4] H. Leng, A. Ohmura, L. N. Anh, F. Ishikawa, T. Naka, Y. K. Huang, A. de Visser, J. Phys.: Condens. Matter **32**, 025603 (2020).