Superconductivity in Ta$_3$Pd$_3$Te$_{14}$ with quasi-one-dimensional PdTe$_2$ chains

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We report bulk superconductivity at 1.0 K in a low-dimensional ternary telluride Ta$_3$Pd$_3$Te$_{14}$ containing edge-sharing PdTe$_2$ chains along crystallographic $b$ axis, similar to the recently discovered superconductor Ta$_4$Pd$_3$Te$_{16}$. The electronic heat capacity data show an obvious anomaly at the transition temperature, which indicates bulk superconductivity. The specific-heat jump is $\Delta C / \gamma (T_c) \approx 1.35$, suggesting a weak coupling scenario. By measuring the low-temperature thermal conductivity, we conclude that Ta$_3$Pd$_3$Te$_{14}$ is very likely a dirty $s$-wave superconductor. The emergence of superconductivity in Ta$_3$Pd$_3$Te$_{14}$ with a lower $T_c$ compared to that of Ta$_4$Pd$_3$Te$_{16}$ may be attributed to the lower density of states.

Superconductivity (SC) in low-dimensional systems attracts sustained attention in SC community. The discovery of first layered cuprate superconductor (La, Ba)$_2$CuO$_4$, has set off a wave of exploring high-$T_c$ superconductors. Since after, a number of new superconductors with low dimensional structures, such as quasi-two-dimensional (Q2D) strontium ruthenate, ferroarsenides, bismuth oxysulfides, quasi-one-dimensional (Q1D) transition-metal chalcogenides, ternary tellurides, and newly discovered chromium-based compounds, were reported to display the features of novel SC. The spin (charge) fluctuations, strong electron-electron correlations, or metal-insulator boundaries among low-dimensional systems constitute the newly strategic prerequisites to explore high-$T_c$ superconductors. Generally, the presence of some transition metal elements among them, which bear strong electron correlations, are believed to play a significant role in producing the exotic pairing glue.

Owing to the inherent nature of transition metal chalcogenides, the low-dimensional structures and rich physical properties, e.g., density-wave instability, thermoelectricity, and SC, are prevalent among them. The tellurides, as compared with sulfides or selenides, are quite special in terms of its structures and properties because of the diffuse nature of the tellurium orbitals, and thus far rarely studied. Recently, we reported the observation of SC with $T_c = 4.6$ K in a ternary telluride Ta$_4$Pd$_3$Te$_{16}$ with Q1D PdTe$_2$ chains. The detailed studies of its pairing symmetry were followed in applying the techniques of scanning tunneling microscopy, low-temperature heat capacity and thermal conductivity. The results indicate an anisotropic gap structure with the possible presence of nodes, although electronic structure calculations show the contributions of Pd 4$d$ electrons to the density of states at Fermi level are pretty small.

From a crystal-structure viewpoint, Ta$_3$Pd$_3$Te$_{14}$ also belongs to a layered compound resulting from the condensation of Pd-based octahedral chains, Ta-based bicapped trigonal prismatic chains, and Ta-based double octahedral chains. If the Ta-based double octahedral chains are replaced by Ta-based single octahedral chains, the condensation of the three different types of chains would form the atomic layer of a new compound Ta$_3$Pd$_3$Te$_{14}$, which was firstly synthesized by Limatta and Ibers in 1980. The major difference between them in structure is well reflected from Fig. 1(f), which shows the projection view of one atomic layer of Ta$_3$Pd$_3$Te$_{14}$ and Ta$_4$Pd$_3$Te$_{16}$ along the $b$ axis. The structural details of Ta$_3$Pd$_3$Te$_{14}$ are discussed below. Then, considering the close structural relationship of this material with the superconductor Ta$_4$Pd$_3$Te$_{16}$, a natural question is whether the former is as well a superconductor.

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In this paper, we report the observation of SC with $T_c = 1.0\,\text{K}$ in layered ternary telluride $\text{Ta}_3\text{Pd}_3\text{Te}_{14}$ with Q1D $\text{PdTe}_2$ chains. The bulk SC was identified by the electronic heat capacity data, which shows an obvious anomaly at the transition temperature. The specific-heat jump $\Delta C/(\gamma n T_c)$ $\approx 1.35$ indicates $\text{Ta}_3\text{Pd}_3\text{Te}_{14}$ may be a weakly coupled superconductor. In addition, the result of low-temperature thermal conductivity measurements of $\text{Ta}_3\text{Pd}_3\text{Te}_{14}$ crystal down to 80 mK suggests a dirty $s$-wave superconducting gap. We summarize our results by discussing the similarities and differences between the closely related superconductors of $\text{Ta}_3\text{Pd}_3\text{Te}_{14}$ and $\text{Ta}_4\text{Pd}_3\text{Te}_{16}$, and compiled an extended list of their physical properties.

**Results**

Single crystals of $\text{Ta}_3\text{Pd}_3\text{Te}_{14}$ were grown using a self-flux method, rather than the vapor transport method previously used$^{21}$. Shiny flattened needle-like crystals with a typical size of $2 \times 0.15 \times 0.1\,\text{mm}^3$ were harvested, as shown in Fig. 1(a). The X-ray diffraction (XRD) pattern at 298 K by a conventional $0-2\theta$ scan for the crystals lying on a sample holder is shown in Fig. 1(b), in which we can observe only multiple peaks arising from the diffraction from $(101)$ planes, consistent with the layered crystal structure of $\text{Ta}_3\text{Pd}_3\text{Te}_{14}$. $\text{Ta}_3\text{Pd}_3\text{Te}_{14}$ crystallizes in space group $\text{P}2_1/m$ with a monoclinic unit cell of $a = 14.088(19)\,\text{Å}$, $b = 3.737(3)\,\text{Å}$, $c = 20.560(19)\,\text{Å}$, and $\beta = 103.73(5)^\circ$ at 123 K$^{21}$. As seen in Fig. 1(d), the layered slabs compose of successively six different chains of three different types. The three types of chains are Ta-based bicapped trigonal prismatic chains, Pd-based octahedral chains, and Ta-based octahedral chains, respectively. The arrangement of the chains, in such a way that every Pd-based chain has two adjacent Ta-based chains and vice versa, constitute the layered slab as clearly depicted in Fig. 1(e). For simplicity, hereafter we define the $a'$ axis as to be parallel to the [101] direction and the $c'$ axis as to be perpendicular to the $(010)$ plane. The interplane spacing at room temperature is determined to be 6.418 Å, and this value is well consistent with the calculated one of 6.397 Å using the above mentioned parameters at 123 K, when taking into account the temperature difference. To compare the obvious difference of the interlayer spacing of $\text{Ta}_3\text{Pd}_3\text{Te}_{14}$ and $\text{Ta}_4\text{Pd}_3\text{Te}_{16}$, we plot the third reflection together, namely $(-606)$ and $(-309)$ peaks, in the inset of Fig. 1(b), from which one can easily find the interplane spacing of $\text{Ta}_3\text{Pd}_3\text{Te}_{14}$ is $\sim 2\%$ smaller than that of $\text{Ta}_4\text{Pd}_3\text{Te}_{16}$.


Figure 2. **Electrical transport and superconducting phase-diagram.** (a) Temperature dependence of electronic resistivity of Ta₃Pd₃Te₁₄ crystal (Sample 1) along the b axis. (b) shows the power-law fit to ρ₀ = ρ₀ + ATⁿ in the data range of 2 and 25 K. (c) zooms into the low-temperature range to clearly show the superconducting transition. (d) The low-temperature resistivity in fields H || cʰ up to 0.1 T, from which the upper critical field (Hc₂) is derived. (e) The red dashed line represents the Werthamer-Helfand-Hohenberg (WHH) fitting. (f) The extracted upper critical field Hc₂ of Ta₃Pd₃Te₁₄ crystal (Sample 2) for different field orientations.

full width at half-maximum is only 0.05°, indicating the high quality of the crystals. The chemical composition determined by an energy-dispersive X-ray spectroscopy (EDS), are collected in a number of crystals, and the average results confirm that composition of the crystals is the stoichiometric Ta₃Pd₃Te₁₄ within the measurement errors. The SEM image, shown in the upper right corner of Fig. 1(c), has a morphology with stripes along the b axis (chain direction), consistent with the preferential crystal growth along the chain direction.

Figure 2(a) shows temperature dependence of electronic resistivity along the b axis (ρ₀) for the Ta₃Pd₃Te₁₄ crystal (Sample 1). The larger room temperature resistivity (1.18 μΩ cm), than that (0.61 μΩ cm) of Ta₄Pd₃Te₁₆, indicates Ta₃Pd₃Te₁₄ is less conductive, consistent with the previous reports₂¹,₂². The temperature dependence of resistivity shows a metallic behavior without any obvious anomaly down to Tc = 1.0 K, at which a sharp superconducting transition appears, as clearly depicted in Fig. 2(c). The value of Tc is 3.6 K less than that of Ta₄Pd₃Te₁₆. The onset, midpoint, and zero-resistance temperatures are 1.02 K, 0.94 K, and 0.81 K, respectively, and the superconducting transition width ΔTc is 0.13 K. The ρ₀(T) data between 2 and 25 K can be well fitted by to ρ₀ = ρ₀ + ATⁿ, giving a residual resistivity ρ₀ = 5.13 μΩ cm and n = 2.83 [Fig. 2(b)]. The value of n more than 2 was also observed in Ta₄Pd₃Te₁₆, which was attributed to the phonon-assisted s-d interband scattering. The residual resistivity ratio (RRR) is estimated to be RRR = ρ₀(300 K)/ρ₀ ~ 23, similar to that of Ta₄Pd₃Te₁₆ (see Table 1).

Figure 2(d) plots the low-temperature resistivity of Ta₃Pd₃Te₁₄ crystal (Sample 1) for H || cʰ up to 0.1 T. Upon increasing the field, the superconducting transition is suppressed to lower temperature. The extracted upper critical fields Hc₂(T) for H || cʰ, determined by using 90% criterion, i.e., the field at which ρ₀ reaches 90% of the normal state resistivity, are shown in Fig. 2(e). We applied the isotropic one-band Werthamer-Helfand-Hohenberg (WHH) formalism to roughly estimate Hc₂. As can be seen in Fig. 2(e), Hc₂ for H || cʰ is estimated to be 0.075 T at zero temperature with the derived Maki parameter α = 1.7 and spin-orbit coupling parameter λso = 1.2. However, by employing the orbital limiting field μ0Hc₂orb = −0.69 μ0μd(Hc₂/ΔTc)Tc = 0.1 T in WHH model and the BCS Pauli-limiting field μ0Hc₂BCS = 1.84Tc = 1.84 T, the Maki parameter α = √μ0Hc₂/ΔTc is calculated to be 0.077. This inconsistence between the calculated value of α and the fitted one may originate from the anisotropic effect in Ta₃Pd₃Te₁₄. The extracted Hc₂ with fields applied along a’, b, and c’ directions for Sample 2 are shown in Fig. 2(f), and the resistivity data of Sample 2 are not shown here. By roughly linear extrapolations, the anisotropic Hc₂ at zero temperature are estimated to be 0.21, 0.27 and 0.086 T for the a’, b and c’ directions. Using the Ginzburg-Landau formula, the superconducting coherence length ξ are calculated to be 545, 703 and 223 Å for a’, b and c’ directions, respectively, which are much larger than those of its analog Ta₄Pd₃Te₁₆. The SC in Ta₃Pd₃Te₁₄ is anisotropic but as well three-dimensional in nature, similar to other superconductors with Q1D
Table 1. Comparison of some physical parameters of the superconductors Ta₃Pd₃Te₁₄ (present work) and Ta₄Pd₃Te₁₆²₀ and Ta₄Pd₃Te₁₆¹⁷,¹⁹,²⁰. RRR, ΔTₚ, ΔH₀, γ, γ₁, λ, λ₁, N₀(E_F), and ΔC/γ₂T_c denote the residual resistivity ratio, superconducting transition temperature, transition width, upper critical field, electronic specific-heat coefficient, Debye temperature, electron-phonon coupling constant, electron-nonphonon coupling constant, density of states at Fermi level, and dimensionless specific-heat jump, respectively.

| Physical parameters | Ta₃Pd₃Te₁₄ | Ta₄Pd₃Te₁₆ |
|---------------------|------------|------------|
| RRR                 | 23         | 26         |
| T_c (K)             | 1.0        | 4.6        |
| ΔT_c (K)            | 0.13       | 0.76       |
| ρ(T)/ρ₀ (Ω cm)      | -0.14      | -0.44      |
| ρₚ(T)/ρₚ₀ (Ω cm)    | 0.075      | 3.3        |
| γₑ (mJ mol⁻¹ K⁻²)   | 28.2       | 46.1       |
| Θ_D (K)             | 151.6      | 148.8      |
| γ₁                  | 0.51       | 0.77       |
| λ₁                   | 1.99       | 1.53       |
| λ₁h                 | 1.53⁶       | 0.12⁶       |
| N₀(E_F) (eV⁻¹ fu⁻¹) | 3.4⁹       | 9.6¹⁹, 8.5¹⁹ |
| ΔC/γ₂T_c            | 1.35       | 1.40       |

Figure 3. Temperature dependence of specific heat. (a) C/T vs T, in which the red dashed line represents the fit with the formula C/T = γₑ + βT² for the normal-state data from 1.2 to 6.5 K. (b) The electronic specific heat divided by temperature Cₑ/T in the superconducting state, where Cₑ/T = C - βT³.

characteristics, e.g., Ta₃Pd₃Te₁₄¹⁷, Nb₃Pd₃Se₇²⁴, and Nb₄Pd₃Se₅²⁵, since the interchain coherence length ξₑ and ξₜₑ are much larger than the distance between two arbitrarily adjacent chains.

The low-temperature specific heat data of Ta₃Pd₃Te₁₄ crystals, plotted as C/T vs T, are shown in Fig. 3(a). We fit the normal-state data from 1.2 to 6.5 K, employing the usual formula C/T = γₑ + βT², which is represented as the red dashed line. The fitting yields an electronic heat capacity coefficient γₑ = 28.2 ± 0.9 mJ mol⁻¹ K⁻², and a phononic coefficient β = 11.14 ± 0.04 mJ mol⁻¹ K⁻⁴. The calculated Debye temperature Θ_D = 151.6 K is close to the value of Ta₄Pd₃Te₁₆ consistent with the fact that the structures of two tellurides are closely related. However, the value of extracted coefficient γₑ is nearly 40% smaller than that of Ta₄Pd₃Te₁₆. Using the relation N₀(E_F) = 3γ₁/κₕ₀²π² for noninteracting electron systems, where κₕ₀ is the Boltzmann constant, we estimated the density of states at the Fermi level N₀(E_F) to be about 11.9 ± 0.8 eV⁻¹ fu⁻¹, which is 3.5 times that of the bare density of states N₀₀(E_F), obtained from the previous band-structure calculations. Therefore, the larger renormalization factor [N₀(E_F)/N₀₀(E_F) = 1 + λ], than that for Ta₄Pd₃Te₁₆ suggests much stronger electron-electron correlations in Ta₃Pd₃Te₁₄, although the recent band-structure calculations are concluded with a higher N₀₀(E_F) = 9.6 eV⁻¹ fu⁻¹ for Ta₄Pd₃Te₁₆ thus resulting in a much lower renormalization factor. To extract the electron-nonphonon coupling strength λ₁ₜₑ in λ, we estimate the electron-phonon coupling constant λ₁ₜₑ by employing the McMillan formula, λ₁ₜₑ = [1.04 + μ l'(θ_D/1.45T_c)]/[1 - 0.62μ l'(θ_D/1.45T_c) - 1.04], where the Coulomb repulsion parameter μ is empirically set to be 0.13. The estimated value of λ₁ₜₑ is 0.51, a little bit smaller than that of the superconductor Ta₄Pd₃Te₁₆. However, the resultant constant λ₁ₜₑ = λ - λ₁ₜₑ = 1.99 is much larger than that of Ta₄Pd₃Te₁₆, possibly indicating much larger electron correlations in the former compound.
Discussion

We discuss the electronic heat capacity $C_{el}$ of Ta$_3$Pd$_3$Te$_{14}$ crystals in low-temperature range, obtained by $C_{el} = C - \beta T$. As can be seen in Fig. 3(b), a characteristic superconducting jump ($\Delta C_{el}$) shows up around ~1 K, confirming the bulk SC. The $\Delta C_{el}/T_c$ is estimated to be 38.0 mJ mol$^{-1}$K$^{-2}$ and the midpoint temperature of the thermodynamic transition is 1.0 K, consistent with the superconducting transition in low-temperature resistivity.

The dimensionless specific-heat jump can be calculated to be 1.35, smaller than the theoretical value (1.43) of the well-known BCS theory, indicating Ta$_3$Pd$_3$Te$_{14}$ may be a weakly coupled superconductor. Unfortunately, due to the insufficient data points, we are unable to fit $\Delta C_{el}(T)$ with standard gap functions to give valuable information about the gap symmetry.

To shed light on the superconducting gap structure, we measured the thermal conductivity of Ta$_3$Pd$_3$Te$_{14}$ single crystal (Sample 2) in zero and magnetic fields (along $c^*$ direction). The dashed lines are fits to the formula $\kappa/T = \kappa_0 + bT$. The black dashed line is the normal-state Wiedemann-Franz law expectation $L_0/\rho_0$. (b) The field dependence of $\kappa/T$ at 0.1 K. (c) Normalized residual linear term $\kappa_0/T$ as a function of normalized field $H/H_c$ for the clean s-wave superconductor Nb$^{36}$, the dirty s-wave superconducting alloy InBi$^{32}$, the multi-band s-wave superconductor NbSe$_2$$^{37}$, and an overdoped d-wave cuprate superconductor Tl-2201$^{38}$.

Figure 4. Low-temperature thermal conductivity data. (a) Low-temperature thermal conductivity of Ta$_3$Pd$_3$Te$_{14}$ crystal (Sample 2) in zero and magnetic fields applied along $c^*$ direction. The dashed lines are fits to the formula $\kappa/T = \kappa_0 + bT$. The black dashed line is the normal-state Wiedemann-Franz law expectation $L_0/\rho_0$. (b) The field dependence of $\kappa/T$ at 0.1 K. (c) Normalized residual linear term $\kappa_0/T$ as a function of normalized field $H/H_c$ for the clean s-wave superconductor Nb$^{36}$, the dirty s-wave superconducting alloy InBi$^{32}$, the multi-band s-wave superconductor NbSe$_2$$^{37}$, and an overdoped d-wave cuprate superconductor Tl-2201$^{38}$.

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of our measurement is not low enough, comparing to the $T_c$. Down to lower temperature, the curve in zero field should deviate from the linear behavior.

Since we can not extrapolate $\kappa_0/T$ at low field, we plot the field dependence of $\kappa/T$ at $T = 0.1 T_c$, well below $T_c$, in Fig. 4(b) to get more information about the superconducting gap structure of Ta$_4$Pd$_3$Te$_{16}$. One can see that the increase of $\kappa/T$ at low field is rather slow, and the curve is similar to that of dirty $s$-wave superconducting alloy InBi$_2$, which is shown in Fig. 4(c). By using the estimated value of the coherence length $\xi$ along $b$ direction, the formula $\xi = 0.18 \hbar v_F/\kappa_0 T_c$ gives the Fermi velocity $v_F = 5.11 \times 10^5$ m s$^{-1}$. Then, according to the relationship $\kappa_0 T_c/\gamma = \Delta/\sqrt{3}$, the electron mean free path is estimated to be $\ell_e = 322 \AA$, which is much smaller than the $b$-direction $\xi$. This result indicates that the $s$-wave superconductor, Ta$_4$Pd$_3$Te$_{14}$, is very likely a dirty $s$-wave superconductor.

It is instructive to compare the physical properties of the two structural closely related compounds of Ta$_3$Pd$_3$Te$_{14}$ and Ta$_4$Pd$_3$Te$_{16}$ which we summarize in Table 1. Both of the two tellurides show Q1D characteristic with Q1D PdTe$_2$ chains. The larger RRR could account for the much sharper superconducting transition for Ta$_3$Pd$_3$Te$_{14}$. Although $T_c$ of Ta$_4$Pd$_3$Te$_{16}$ is 3.6 $K$ less than that of Ta$_3$Pd$_3$Te$_{14}$, the former compound show much stronger electron correlations, verified by the larger renormalization factor and larger electron-nonphonon coupling strength $\lambda_{nnp}$. The small values of both $\Delta C(\gamma T_c)$ and $\lambda_{nnp}$ indicate Ta$_4$Pd$_3$Te$_{16}$ is a weakly coupled superconductor. In addition, if assuming the Drude model, in which the electron-electron interactions are neglected, is applicable, a superconductor is expected to have a lower $T_c$ for a lower value of Sommerfeld coefficient $\gamma T_c$, which in this case signifies the lower density of states at Fermi level. This simple conclusion is compatible with the general trend for the above two superconductors. Therefore, the lower $T_c$ in the title compound, may be attributed to the lower density of states at Fermi level. In this sense, by tuning the Fermi level of Ta$_3$Pd$_3$Te$_{14}$ or Ta$_4$Pd$_3$Te$_{16}$ by the way of doping or proper intercalations, the value of $T_c$ may be enhanced. By the way, in recently discovered PdTe$_2$ or PdS chains based superconductors, there have been several pieces of work that show the evidences of two-gap SC$^{17,34,35}$. However, our results presented above indicate the new superconductor Ta$_4$Pd$_3$Te$_{16}$ with PdTe$_2$ chains is very likely a fully gapped $s$-wave one. We have previously reported that Ta$_3$Pd$_3$Te$_{14}$ is possibly a two-gap superconductor with a gap symmetry of $s + \delta$ waves$^{17}$. Thus, if assuming the reduced part of electronic states at the Fermi level in Ta$_3$Pd$_3$Te$_{14}$ compared to that in Ta$_4$Pd$_3$Te$_{16}$, is primarily due to the reduced contribution from the Pd $d$ states, it would be reasonable to see only a $s$-wave gap left in the former compound.

Methods

Powders of the elements Ta (99.97%), Pd (99.995%) and Te (99.999%) with a ratio of Ta: Pd: Te = 2: 3: 10 were thoroughly mixed together, loaded, and sealed into an evacuated quartz ampoule. The sample-loaded quartz ampoule is then heated to 1223 $K$, held for 24 h, and cooled to 723 $K$ at a rate of 5 $K/h$, followed by furnace cooling to room temperature. The above procedures are similar to that in growing Ta$_4$Pd$_3$Te$_{16}$ crystals$^{6}$. The chemical composition is checked by an EDS with an AMETEK EDAX (Model Octane Plus) spectrometer, equipped in a $\delta$-wave gap left in the former compound.

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Author Contributions
W.H.J., Y.L. and C.H.Z. synthesized the crystals of Ta₃Pd₃Te₁₄. W.H.J., X.F.X., Y.K.L. and N.Z. performed transport measurements and heat-capacity measurement. The low-temperature thermal conductivity measurements were performed by L.P.H. W.H.J., S.Y.L., G.H.C. and Z.A.X. analyzed the whole data. W.H.J. and S.Y.L. wrote the main manuscript text. All authors reviewed the manuscript.

Additional Information
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